

**AN URBAN AIR QUALITY MODEL  
FOR THE CONTROL OF  
PARTICULATE POLLUTION**

Gerald H. Miles

BSc (Griffith), Dip. Community Sc.(Murdoch)

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## Declaration

Except where otherwise indicated  
this thesis is my own work.

A handwritten signature in cursive script, appearing to read "G. Miles".

Gerald H. Miles  
November 1987

## Preface

The following papers have been accepted or submitted for publication from the research undertaken towards this thesis:

Simpson R.W., Miles G.H (1987) Controlling emissions to avoid violations of health standards for short and long term exposures to TSP concentrations. *Accepted for Atmospheric Environment*

Simpson R.W., Miles G.H., Littleboy M., Verrall K. (1987) The Brisbane TSP Study: controlling for the effects of long term exposure. *Accepted for publication Clean Air.*

Miles G.H., Simpson R.W. (1987) The Brisbane TSP study: controlling for the effects of short term exposure. *Accepted for publication Clean Air.*

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Miles G.H., Simpson R.W., Glikson M. (1987) The Brisbane TSP study: Part III toward an emissions control strategy for inhalable and respirable particulates. *Submitted for publication Clean Air.*

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## Abstract

A model, the Total Suspended Particulate (TSP) Hybrid Emissions Control (HEC) model, has been developed for the management of particulate air pollution in the Brisbane urban area. This model, based on deterministic, statistical and hybrid modelling approaches, has enabled predictions of ambient TSP concentrations to be related to both the violation of a number of air quality goals and the specific emissions that have contributed to those violations.

Violations of the annual arithmetic mean goal for long term exposure to TSP are estimated using the Atmospheric Turbulence Diffusion Laboratory (ATDL) model. Once calibrated for background crustal TSP, the ATDL model predicts annual arithmetic mean TSP to within a factor of two of observed TSP concentrations. To examine violations of goals and appropriate control strategies for acceptable short term exposure to TSP, the output of the calibrated ATDL model is linked to the results of a "hybrid" model. This model combines deterministic and statistical techniques to estimate the frequency distribution for a given pollutant. From the "hybrid" approach the two parameter lognormal distribution has been found to be appropriate for annual TSP data sets in Brisbane. Using wind speed data to provide a measure of the variation for this distribution (ie the shape parameter) the ATDL model output is then related to an entire frequency distribution for predicted particulate concentrations. This allows emission controls to be linked through the ATDL model to violations of both long and short term exposure standards. The estimates of 24 hour maxima using this technique have been found to be generally within a factor of two of expected maxima for TSP. The uncertainty in assuming a lognormal distribution has been estimated and found to lead to a conservative control strategy. Similarly, the use of annual wind speed data as a measure of variation for TSP data will result in over-estimation of TSP maxima. It appears that this technique is applicable to current management strategies based on the 1978/79 particulate emissions inventory for the Brisbane urban area and, by examining the two primary assumptions of the model, the technique may be applicable to other urban airsheds (eg Sydney and Melbourne). It may also be possible to use the HEC approach for the control of inhalable and respirable particles.

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# Chapter 1

## INTRODUCTION

Mathematical models can be used in a variety of ways to assess existing conditions and effects that are likely to occur as a result of present or future emissions of air pollutants (Stern et al, 1984). These models play an important role in the development and implementation of control strategies for the management of urban air quality by linking particular emissions to ambient concentrations that may affect the urban community. Further effort is required, however, to improve the accuracy of air quality models and the input data they require (Hayes and Moore, 1986) if the results of these models are to be applied effectively to the control of air pollution. This is reinforced by the fact that control measures introduced to improve air quality can have significant social and economic effects (Dennis et al, 1983). If the aim is to regulate and manage atmospheric pollution, then it is also desirable that the output from these models yields information relevant to definitions of acceptable air quality.

Different modelling approaches satisfy these requirements for accuracy and relevance with varying degrees of success. The merits of these approaches will be discussed in the following section. The air quality standards and goals, officially recognised by government control agencies as the most appropriate to define clean air, will also vary greatly (Bentley et al, 1987). The standards that are appropriate for this study will therefore also be discussed (see section 1.2). An outline of the thesis will then be presented.

Although the term air quality refers to the full mix of compounds we see and breathe, the management of this resource is commonly in terms of its components. This is reflected in the way air quality health standards are set based on health and damage criteria for individual pollutants (Bentley et al, 1987). For this study particulate air pollution has been chosen as the component of interest because it has been identified as an ongoing concern in Australia (Department of Arts, Heritage and Environment, 1985) and also represents a significant health concern throughout the world (Suess and Craxford, 1976). For example, within Australia particulate concentrations have been in excess of defined standards have occurred in

Brisbane (Division of Noise Abatement and Air Pollution Control, 1986) and this urban area has been selected as the focus for the study. The specific characteristics and health effects associated with airborne particulate matter that warrant this concern are outlined in Chapter 2.

## 1.1. MODELLING APPROACHES

The development of air quality models can be categorised into the two fundamental approaches of deterministic and statistical modelling. These approaches are well reviewed elsewhere (Benarie, 1980, Stern et al, 1984, Taylor, 1985, Jakeman et al, 1986a, Jakeman et al, 1987a) and consequently only a general discussion of the merits and objectives of each approach that are relevant to particulate air pollution in an urban environment will be presented here. The hybrid modelling approach will also be briefly introduced as it combines useful facets of both deterministic and statistical approaches. This will provide some context and rationale for the modelling approach adopted in this study.

### 1.1.1. Deterministic Models

Deterministic models seek to establish a causal link between the pollutant emitted and the ground level concentration that may be expected after atmospheric transport and dispersion. Spatial and temporal variation in concentrations may then be predicted depending on the model used. Through this approach it is possible both to regulate the emissions from specific sources, thereby maintaining acceptable ambient concentrations, and to obtain further insights into the physical processes involved in air pollution episodes. It is noted, however, that this link is only adequately generated by deterministic models for concentrations that occur most frequently; the prediction of more extreme events, such as maximum concentrations, can be quite poor (Jakeman et al, 1987a). Large errors in the estimates of these high concentration air pollution episodes result primarily from the turbulent and stochastic nature of atmospheric diffusion (Fox, 1981, Pasquill and Smith, 1983) currently not accounted for in deterministic models.

The specific deterministic models that have been constructed for particulate and inert pollutants vary greatly in their detail and are too numerous to review individually. They can, however, be discussed in terms of some common techniques.

Box models may be used as first approximations of pollutant concentrations for a defined volume of air. By assuming that uniform mixing exists the following equation (1.1) (Stern et al, 1984) can be used to estimate the steady state

concentration,  $\chi$ , within a box or volume of air defined by a given distance,  $\delta x$ , and mixing height,  $L$ , for an area emission rate,  $q_a$ , and mean wind speed,  $u$ .

$$\chi = \delta x q_a / (L u) \quad (1.1)$$

Gaussian modelling techniques have been used for many years and produce reasonable estimates of ground level concentrations for inert pollutants up to 20km from individual point sources (eg see Stern et al, 1984). The technique has also been adapted to predict concentrations from line sources. It assumes that continuous emissions are dispersed at a rate inversely proportional to wind speed and that a Gaussian distribution can be used to represent concentrations along vertical and horizontal axes through the plume.

Other more complex techniques, such as gradient transport models, can be used for continuous line and area sources. These models calculate the rate of diffusion for a given emission, assuming that the turbulent flux is proportional to the gradient of concentration. Care must be taken, however, that the numerical instability resulting from the random errors in successive iterations (Stern et al, 1984) can be overcome.

Trajectory models are commonly used for single parcels of air moving along a given path. As the parcel moves it can chemically react (not included in the consideration of particulate modelling), be added to, and diffuse according to inputs of emissions, wind speed and wind direction. Again this is well suited to predicting the effects of single source emissions but to manage urban air it is important to predict concentrations that may result from multiple sources over a large area. In order to make these predictions models based upon multiple emissions strengths, that may incorporate a number of techniques to account for both point and area sources, must be used.

While the source conditions are very complex within an urban area the ground level concentrations of pollutants will tend to vary less than under a single source scenario (Stern et al, 1984). This has probably contributed to the relative success of simple models used to predict particulate air pollution in urban areas. For example, the Atmospheric Turbulence Diffusion Laboratory (ATDL) model, based on a simple "reciprocal plume" concept for area source dispersion and supplemented by Gaussian plume dispersion techniques for point sources (Hanna, 1972), has been found to produce results comparable to the more complicated Climatological Dispersion Model (CDM) (Turner et al, 1972). The performance of the ATDL model relative to other more complicated models has also been found to be adequate given the uncertainties involved in data collection (Hanna, 1971).

Simple models, such as the ATDL model, offer advantages to air quality management through the general requirement for less data. This allows the simple model to be more readily validated and calibrated without adding extensively to the monitoring requirements in a given airshed. This must be balanced against requirements for information regarding the atmospheric processes involved. Simple models, by definition, provide less explanatory detail than models involving more complex interactions between a larger number of variables.

### 1.1.2. Statistical Distribution Models

Guidelines for acceptable air quality are commonly quoted as statistics of a given frequency distribution, such as the mean, median, maximum and 98<sup>th</sup> percentile. Statistical distribution models link ambient concentrations and the violation of prescribed goals through estimating the frequency distribution of air pollution concentrations for a given averaging time (Larsen, 1969, Georgopolous and Seinfeld, 1982, Jakeman et al, 1987a). The appropriate statistic can then be selected from this distribution for comparison with prescribed guidelines for acceptable levels of air pollution. Although there is no *a priori* rationale for suggesting that air pollutant concentrations are likely to be defined by a particular distribution (Georgopolous and Seinfeld, 1982) evidence is mounting that certain conditions and pollutants may have distributions in common.

The types of statistical distribution models that have been found to be appropriate for air pollution data include the exponential and the two parameter lognormal, Weibull and gamma distributions (Larsen, 1969, Simpson et al, 1986, Jakeman et al, 1986b, Jakeman et al, 1987a). Three parameter distributions of lognormal, Weibull and gamma have also been found to be useful in representing air pollution data (Georgopolous and Seinfeld, 1982).

Statistical models are designed to predict all events well. Alone, however, they do not provide a causal link between the violations of a standard and the physical processes that have produced this unacceptable concentration. They provide temporal and some limited spatial information but this can only be used in relation to the data that have been collected and a time series cannot be developed (Jakeman et al, 1987a, Bai Jun et al, 1987). Hence the action taken (eg emissions reductions of specific sources under certain conditions) to achieve acceptable air quality based upon this approach can be limited.

Hayes and Moore (1986) suggest that if any model is to be used to assist decision making then the uncertainties in the model predictions must be

characterised. This uncertainty has an irreducible part due to the stochastic nature of atmospheric processes and a reducible part due to imperfect model formulation. While the uncertainty of predictions is often not characterised for deterministic models, statistical models can readily quantify these errors (Jakeman et al, 1987a). Of course errors may arise if assumptions necessary for the application of statistical techniques are invalid. Although air pollutant concentrations are inherently random variables (Georgopolous and Seinfeld, 1982) there are conditions where data are not independently and identically distributed. At low averaging times (eg minutes) data may be autocorrelated and over certain periods (eg annual) may not be stationary (ie vary seasonally) (Chock, 1982).

Receptor models should be briefly mentioned as they utilise statistical techniques to apportion source contributions to measured ambient concentrations. They are distinct from statistical distribution models as they require knowledge of source characteristics and assumptions of linearity in the contributions of multiple sources to a given concentration. While they do have useful attributes, such as estimating contributions from background sources (see Chapter 4), they are limited in their predictive capabilities and are considered to be of value primarily for interpretation (Batterman et al, 1984).

### 1.1.3. Hybrid Models

Both deterministic and statistical methods, therefore, have important benefits and limitations in the application of the respective models to given management situations. For urban air pollution deterministic and statistical techniques have been combined to produce a hybrid modelling approach that links entire frequency distributions to total emissions (Simpson et al, 1986, Jakeman et al, 1987a). This approach has been developed over recent years at the Centre for Resource and Environmental Studies at the Australian National University (Simpson et al, 1983, Simpson and Jakeman, 1985, Jakeman and Taylor, 1985, Jakeman et al, 1985, Taylor, 1985, Simpson et al, 1986, Simpson et al, 1987). Table 1-1 outlines the applications of this approach to a variety of source types and data sets.

Here it is proposed that the causal relationship be established through deterministic techniques and used to predict concentrations, over the deterministic model's most reliable range, that are within acceptable levels of accuracy. The statistical submodel is then used to relate these values to the parameters of a frequency distribution from which confidence limits can be calculated and used to quantify uncertainty in the predictions. As mentioned above, hybrid models developed for urban areas have made predictions that relate to total emissions

**Table 1-1:** Applications of the CRES hybrid models\*;  
from Jakeman et al, 1987a

Source Type	Deterministic Model	Statistical Model	Pollutant Data Sets	References
AREA	ATDL-related	gamma	10 years of 24h average acid gas at 3 sites, 9 years at 1 site, 8 years at 2 sites in Newcastle	Jakeman and Taylor (1985)
	ATDL-related	lognormal	1 year of 1h average CO at 1 site in Canberra	Simpson, Daly and Jakeman (1983)
	ATDL-related	lognormal	1 year of TSP at 3 sites in Brisbane	Simpson, Daly and Jakeman (1983)
	ATDL-related	lognormal	6 years of TSP at 6 sites and 6 years of nephelometer data at 2 sites in Brisbane	Simpson et al (1986)
POINT	Gaussian plume with plume rise	exponential	2 years of 3h average SO <sub>2</sub> at 3 sites in the Upper Hunter Valley	Jakeman and Simpson (1985)
	Gaussian plume	Weibull	2 years of 24h, 8h, 3h, 1h and .5h averages SO <sub>2</sub> at 2 sites in Kalgoorlie	Taylor, Simpson and Jakeman (1986)
LINE	GM Gaussian line source	Weibull	4 months of 1h average CO at 2 sites in Melbourne	Taylor, Simpson and Jakeman (1986)

\*all component statistical distribution models have two parameters (Simpson et al, 1986, Simpson et al, 1987). To control specific sources, however, that contribute to the total emissions within a given airshed, the link between emissions control and violations of standards needs to be refined, especially for the short term exposure standards. The urban air quality model detailed in this thesis (see Chapter 3) attempts to address this need to relate specific source emissions to violations of standards for airborne particulate matter.

## 1.2. APPROPRIATE STANDARDS

Ambient air quality standards are an integral part of any management approach to air pollution control (Prinz, 1976). The standards that are appropriate for the management of particulate air pollution in a given urban area are prescribed by the control authority with relevant jurisdiction. Within Australia these authorities are State government or Territory organisations which function relatively independently of each other. In most Australian States, with the notable exception of Victoria, ambient air quality goals that are not supported by legislative powers

are used as guidelines, as opposed to standards, for acceptable air quality. In producing a model for air quality management, therefore, these goals are used here to determine the appropriate form for the model output.

To adequately protect human health from both acute and chronic effects of air pollution more than one goal tends to be reviewed (Bentley et al, 1987). The goals relating to acute effects refer to short term exposures, such as maximum concentrations that occur less frequently, whereas, the goals relating to chronic effects refer to more frequent mean or median concentrations that are measures of long term exposure. It would, therefore, be an advantage to construct models that could relate emissions to a wide range of standards or goals.

The air quality guidelines, while providing the focus for any air quality management model, may not necessarily be the best measure of potential health effects of a given pollutant. This is due to the lag time between scientific research and the adoption of standards and the standard setting process in which documented scientific evidence is considered along with social, political, economic and cultural determinants. This lag time is inherent in the time taken for research to be properly refereed, published and digested by appropriate structures, such as the National Health and Medical Research Council (NHMRC) in Australia (eg see Bentley et al 1987). In assessing the relevance of the model developed for this purpose it will, therefore, also be necessary to examine current information regarding the health effects of the pollutant under investigation. This assessment will be in addition to that relating to the goals presently adopted for the management of particulate air pollution in Brisbane.

### **1.3. THESIS OUTLINE**

The primary aim of this study is to produce a model that can be used for the management of particulate air pollution; a model relating directly to air quality goals for particulates and one able to facilitate appropriate controls for offending sources and identify areas where additional sources of particulate emissions may produce unacceptable air quality. As the model results hinge upon prescribed goals it will also be necessary to examine the assumptions upon which these are based. This will enable the model to be used with appropriate caution.

The approach used to develop an appropriate model for this purpose is outlined in Chapter 3. This Chapter also contains details of the study area selected, Brisbane, and the available emissions, meteorological and ambient particulate data.

Chapter 4 examines how the effects of long term exposure can be controlled

through the application of an existing deterministic model. This model has required calibration to account for background crustal particulates and has thereby highlighted the importance of determining airborne crustal matter to the management of particulate air pollution. Controlling for the effects of short term exposure is detailed in Chapter 5. Here the results of the deterministic model from the previous Chapter are combined with a hybrid model and the link between specific emissions and an appropriate frequency distribution for the pollutant is made. This allows for the prediction of extreme events associated with short term exposure. Both Chapters 4 and 5 briefly examine how point and area source contributions can be distinguished to allow for appropriate control measures to be adopted and the effects of likely variations in emissions, such as the introduction of unleaded petrol, are also reviewed.

It is clear that some measure of the uncertainties associated with the model predictions should be provided. Uncertainties that can be quantified as a result of the statistical analysis of the data will, therefore, be presented in Chapter 6. This Chapter will also examine the general applicability of the model to other urban areas in Australia. The health relevance of the model must also be reviewed in light of the lag between prescribed goals and the current understanding of the effects of particulate air pollution. This review is outlined in Chapter 7 with the conclusions to this study presented in Chapter 8.



## Chapter 2

# AIRBORNE PARTICULATE MATTER

Fundamental physical and chemical characteristics of particulate air pollution are well described elsewhere (Lodge et al, 1981, Subcommittee on Airborne Particles, 1979). The background information in this section will, therefore, be of specific relevance to the development and application of the model and approach to be proposed for the management of particulate air pollution in Brisbane. This will include, first, some important properties and definitions of terms that aid presentation and understanding in the discussion to follow. The specific health concerns associated with particulates will then be examined. Finally the sources of particulates, their complexity, dispersion and controls will be addressed.

### 2.1. PROPERTIES AND DEFINITIONS

The definitions for particle size that will be used in this discussion and throughout the thesis are:

- *coarse particles* - these are particles greater than  $10\mu\text{m}$  that are ingested in limited quantities and, if ingested, are predominantly confined to the extrathoracic<sup>1</sup> region of the body;
- *inhalable particles* - these particles, less than  $10\mu\text{m}$ , dominate the thoracic<sup>2</sup> fraction with approximately 80% of these particles, between  $2.5$  and  $10\mu\text{m}$ , being deposited in the tracheobronchial region of the lung;
- *respirable particles* - these particles less than  $2.5\mu\text{m}$  will penetrate the alveolar region of the lung with retention times commonly much greater than the two previous size ranges.

The sum weight of these particles within a given volume of air is commonly referred to as the concentration of Total Suspended Particulates (TSP) in  $\mu\text{gm}^{-3}$ . The definitions above are based on the percentage deposition of particles, and therefore the potential health effects of these particles, within the human body

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<sup>1</sup>The extrathoracic region is defined by the nose, mouth and larynx of the human body

<sup>2</sup>The thoracic region is defined by the trachea, bronchia and alveolar section of the human lung

(International Standards Organisation, 1983, Vate et al, 1986, Subcommittee on Airborne Particles, 1979), rather than any other physical or chemical property. The boundaries of these size ranges should be considered as 50% effective cut-points (eg for coarse particles an average mouth breathing subject is expected to inhale and have deposited 50% of particles with a mass mean aerodynamic diameter of  $10\mu\text{m}$ ). It is acknowledged that much larger particles upwards of  $\approx 50\mu\text{m}$  may be inhaled. Due to their proportionately low percentage as deposited particles in the extrathoracic region of the body, however, these larger particles are primarily of concern in high concentration environments (eg occupational exposure).

Airborne particles behave differently depending largely upon their size, shape and chemical composition. Even without anthropogenic emissions the combinations and concentrations of particles in all size classes in the atmosphere are continually changing. Gas phase molecules condense to form larger products which in turn become larger through coagulation and agglomeration until they are more readily removed from the atmosphere by washout and sedimentation. The number of particles will vary in different size classes or at different stages in this cycle with the largest number normally less than  $0.01\mu\text{m}$ . The surface area, however, is dominated by those particles between  $0.1$  and  $1\mu\text{m}$ . Particles in this size range therefore provide a higher proportion of sites for interaction with other compounds. By volume or mass the distribution of particles within size classes is bimodal. One peak occurs at approximately  $0.2\mu\text{m}$  and another at approximately  $10.0\mu\text{m}$  (Subcommittee on Airborne Particles, 1979).

The chemical properties of particles also vary with particle size. Inhalable and respirable particles tend to be chemically dominated by sulphates, nitrates, organic compounds, ammonia and lead whereas coarse particles are commonly characterised by silica (Si), aluminium (Al) and iron (Fe) from soils (commonly referred to as crustal material), sea salt and plant particles (Stern et al, 1984). This will vary with location. The modes of transportation for particles are also size dependent. Coarse particles commonly disperse over short distances unless injected high into the troposphere (Subcommittee on Airborne Particles, 1979) whereas inhalable and respirable particles are dispersed over much greater distances by eddy diffusion and advection. Their rates of removal through sedimentation are much less than for coarse particles with particles less than  $1\mu\text{m}$  having sedimentation velocities of the order of  $<10^{-2}\text{cms}^{-1}$  (Subcommittee on Airborne Particles, 1979).

The optical properties of atmospheric particles should also be considered as these relate to how air quality is perceived (Malm et al, 1980). Visibility is reduced by particles in the atmosphere which increase the extinction coefficient of light,  $b_{ext}$ , and thereby reduce the light intensity,  $-dI$ , according to equation (2.1).

$$-dI = b_{ext} I dx \quad (2.1)$$

where  $I$  is the original intensity of the light and  $dx$  is the path length for the beam (Stern et al, 1984). The extinction of light can be further defined by equation (2.2).

$$b_{ext} = b_{rg} + b_{ag} + b_{scat} + b_{ap} \quad (2.2)$$

where  $b_{rg}$  is the scattering of light due to gases,  $b_{ag}$  is the absorption of light due to gases,  $b_{scat}$  is the scattering of light due to particles and  $b_{ap}$  is the absorption of light due to particles. This equation is dominated by  $b_{scat}$  which is in turn dominated by particles less than  $1\mu m$  (Subcommittee on Airborne Particles, 1979). Other important factors in this equation are chemical composition, moisture content and particle shape.

It is evident that the properties and processes associated with respirable, and to some extent inhalable, particles are quite distinct from those of coarse particles. Indeed it has been suggested that they are almost entirely independent. They, therefore, require different control measures and that the effects of each group can be segregated (Subcommittee on Airborne Particles, 1979). This is examined in the following brief summary of health concerns associated with particulate air pollution.

## 2.2. THE HEALTH CONCERNS OF PARTICULATE MATTER

The primary concerns for human health associated with particulate air pollution are derived mainly from the effects (1) after ingestion (assuming particles must be deposited to have a significant direct physiological effect); and, (2) to visibility (Subcommittee on Airborne Particles, 1979). As outlined in the definitions above, section 2.1, it is predominantly particles less than  $10\mu m$  that are ingested (ie inhaled and deposited) rather than the coarse particles which tend to dominate the gravimetric analysis of TSP.

Of these inhalable particles it is those less than  $2.5\mu m$  that dominate deposition in the alveolar regions of the lung (International Standards Organisation, 1983). Due to the light scattering properties of these respirable particles they also dominate the visibility reducing fraction of airborne particulate matter. Reduced visibility is a source of concern and complaints as it affects the perception of how clean or polluted an environment has become. This will not be dealt with in this thesis, however, it is noted that these aesthetic and psychological effects should not be ignored in assessing the total health effects of air pollution (Takano, 1983).

After ingestion the chemical composition of particles must be taken into account

as composition, in addition to deposition, has been found to be size dependent (Dzubay and Stevens, 1975, Katz, 1980). Factors such as particle retention time in the body and solubility of the particles are also important, with coarser inhalable particles having low retention times and finer, respirable particles, having much greater retention times in the lung. Solubility of particles affects the rate at which their constituents can be absorbed into the blood stream, transported throughout the body and excreted (Subcommittee on Airborne Particles, 1979).

From a recent review of research needs for all priority pollutants in Australia (Murray et al, 1987) it is clear that there is sufficient evidence to warrant concern about, and further research into, the health effects of particulate air pollution as a single pollutant and in combination with other pollutants. This study identifies, from the criteria documents produced by the Air Quality Committee of the National Health and Medical Research Council, that specific health concerns exist for a number of substances including lead and sulphur-particulate compounds. As particulate matter is commonly a mix of different compounds there is also concern that more complicated additive or synergistic effects may occur. Inert particles predominantly less than  $10\mu\text{m}$ , typical of crustal particulate matter, have been found to produce little effect on human health under conditions of controlled laboratory exposure (Subcommittee on Airborne Particles, 1979).

From the perspective of emissions control to protect human health it would be beneficial to distinguish between inhalable and non-inhalable particles. It has been found that TSP tends to be dominated by crustal material which is commonly larger than  $10\mu\text{m}$  (Vate et al, 1986). This material is also predominantly from non-anthropogenic sources (Subcommittee on Airborne Particles, 1979) and must be acknowledged as such in a control strategy for industrial or other anthropogenic sources. If the contribution from crustal sources cannot be resolved from anthropogenic sources then the question must be asked, what are the appropriate levels of control that should be applied to anthropogenic sources if an ambient air quality goal for TSP is exceeded. It is clear that if appropriate relationships are to be identified between airborne particles and their health effects, and the correct sources of these particles are to be effectively controlled, then it is necessary to investigate in more detail the size range and composition of those particles that are inhaled.

## 2.3. SOURCES AND CONTROLS

This section will briefly outline some important considerations for the identification of particulate sources and their controls relevant to this study. As has been indicated coarse particle sources can be both natural and anthropogenic and arise generally from mechanical processes. Inhalable and respirable particles on the other hand tend to be generated from natural and anthropogenic combustion processes and through condensation of gas phase compounds within the atmosphere. Airborne particles within urban atmospheres tend to be dominated by those generated from anthropogenic sources (Subcommittee on Airborne Particles, 1979), however, this should be confirmed before any control measures are taken.

The types of anthropogenic sources that commonly contribute to airborne particulate matter in urban areas include motor vehicles, coal fired power stations, home heating, incineration, open burning, coal fired boilers, quarrying and earth works (Stern et al, 1984). To control any source, natural or anthropogenic, Stern et al (1984) have outlined three basic mechanisms:

1. the application of abatement devices to reduce emissions at the source;
2. alter the raw materials that essentially fuel the pollution; and/or,
3. modify the process to eliminate the offending components.

To select the most appropriate action, however, it is necessary to be able to distinguish between certain types of sources. For example, natural sources such as wind erosion from exposed soil may require changes to land management practices, whereas, particulate pollution from stock piles of similar material may be more appropriately controlled through covering, screening or damping with moisture. More importantly if the natural emissions are unknowingly contributing to monitored ambient air quality, yet as outlined in the previous section they are of little health concern, then unnecessary over-control of anthropogenic sources may occur.

Similarly it is important to be able to identify point from area sources and their relative contributions to ambient air quality. In Australia the community is responsible for a large proportion of particulate emissions from motor vehicles and other domestic sources such as backyard burning. These commonly classes as area sources as opposed to the elevated point sources of private and government run industrial premises. The power to control these different source types is derived from legislation which, in Australia, is spread between the State and Federal governments. Point sources are commonly under the jurisdiction of the relevant

State organisations. Emissions from vehicles, however, are regulated under the Australian Design Rules prescribed by the Federal parliament. This fragmentation of control may pose problems for the air quality management of a given urban area.

Assuming that for already established transport systems the control of vehicle emissions by State pollution control authorities is limited, then the effects of known variations in emissions, such as the introduction of unleaded petrol should be reviewed. This is also important given that vehicle emissions are likely to contribute significantly to the particulate concentrations in a defined urban airshed as they do in Brisbane (Borowski, 1984).

## Chapter 3

# METHODS, STUDY AREA AND DATA

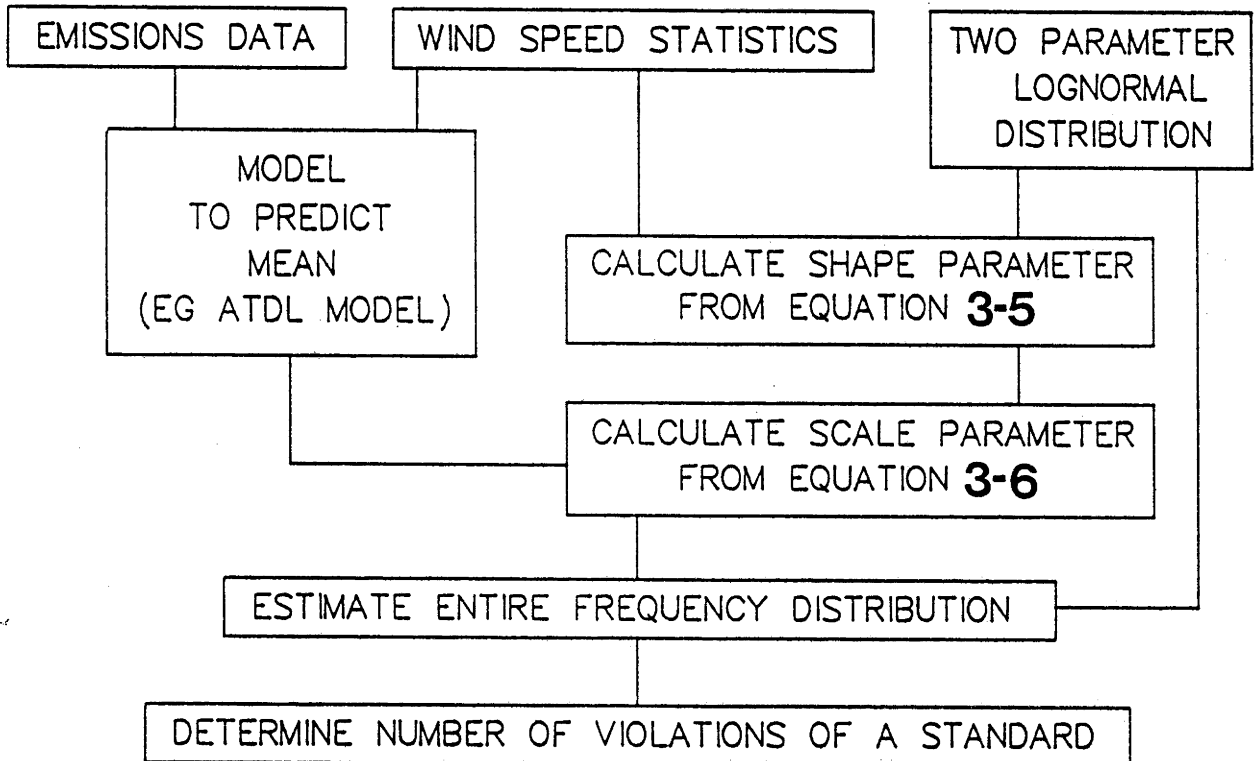
### 3.1. THE MODELLING APPROACH

The development of an appropriate approach to the problem of modelling particulate air pollution in Brisbane has been based upon two main objectives. Firstly, the model output must relate directly to existing air quality goals and, secondly, a link between ambient concentrations and emissions must be made to enable adequate controls to be implemented. In order to satisfy these objectives the hybrid emissions control (HEC) approach has been developed. The approach has been derived from a combination of existing deterministic, statistical and hybrid methods for the modelling of atmospheric pollutant concentrations.

The TSP HEC model developed from this general hybrid approach is described in Figure 3-1. This approach uses a deterministic model based on emissions and meteorological data, such as the Atmospheric Turbulence Diffusion Laboratory (ATDL) model, to predict the annual arithmetic mean for TSP. The lognormal distribution is assumed to be the most appropriate description of annual data sets for the pollutant under investigation. The shape parameter for this distribution is then estimated using an empirical relationship between ground level concentrations and wind speed statistics. This parameter is used with the annual arithmetic mean predicted by the ATDL model to estimate the scale parameter for the lognormal distribution. With both parameters calculated it is then possible to estimate the entire frequency distribution of pollutant concentrations and hence the number of violations of a given standard. Any model if appropriately validated, not just the ATDL model, may be used to make the necessary link between the estimated frequency distribution and emissions. Similarly, the empirical relationship used to estimate the shape parameter may vary. It may be possible to generalise this approach further to include the selection of any frequency distribution following already established methods (Jakeman et al, 1987a).

The ATDL model (Hanna, 1972) has been selected for use in Brisbane to predict

**Figure 3-1:** Diagram of the major steps in developing the TSP hybrid emissions control model



the annual arithmetic mean ground level concentrations of TSP as it is simple, is designed to account for multiple sources, and has been shown to be valid for inert pollutants at a number of urban areas in both the United States of America and Australia (Hanna, 1971, Gifford and Hanna, 1973). It is believed that given adequate emissions and meteorological data the ATDL model may be easily applied and calibrated for many urban areas. If the ATDL model is inappropriate then another deterministic model may be used to predict the annual arithmetic mean based on emissions and meteorological data.

To extend the ATDL model estimates of ground level concentration and produce



the entire distribution of concentrations for the same averaging time the hybrid modelling approach (Jakeman et al, 1987a) has been selected. The method allows accurate estimates of pollutant concentrations, and associated confidence intervals, to be made so that these may be easily related to set standards and control strategies. While a simple statistical distribution model could be used for this purpose the hybrid model provides the link between the frequency distribution and other causal factors such as wind speed. Previous research has also found that a hybrid model can produce acceptable estimates of TSP concentrations in Brisbane (Simpson et al, 1986, Simpson et al, 1987).

### 3.1.1. The ATDL Model

The simple ATDL model is a "physically realistic model" (Hanna, 1971) of use in estimating the ambient concentrations of area source pollution. It has been found to compare well with other more complex models (Benarie, 1978, Gifford, 1974, Gifford, 1972, Hanna, 1971) and in its simplest form:

$$\chi_A = \frac{C Q_A}{u} \quad (3.1)$$

where  $\chi_A (\mu\text{gm}^{-3})$  is the average ground level concentration,  $C$  is a dimensionless constant of atmospheric stability,  $Q_A (\mu\text{gm}^{-2}\text{s}^{-1})$  is the area source strength, and  $u (\text{ms}^{-1})$  is the average windspeed. In applying this model to a given urban area with multiple sources and varying source types the calculations have become slightly more detailed and Hanna (1971), for convenience, categorises emissions into either point or area sources.

For large urban areas it is necessary to integrate equation (3.1) over a number of component areas to better describe the surface concentrations resulting from area sources. The following equation (3.2) presents a relatively simple method for calculating this and includes dispersion effects due to wind direction.

$$\chi = \sqrt{2/\pi} \frac{(d/2)^{1-b}}{ua(1-b)} \{Q_A(0,0) + \sum_{i=-4}^4 \sum_{j=-4}^4 Q_A(i,j) f(i,j) ((2r+1)^{1-b} - (2r-1)^{1-b})\} \quad (3.2)$$

where  $\chi$  is the ground level concentration ( $\mu\text{gm}^{-3}$ ),  $Q_A(i,j)$  is the area source emission for grid  $(i,j)$  ( $\mu\text{gm}^{-2}\text{s}^{-1}$ ),  $u$  is the wind speed ( $\text{ms}^{-1}$ ),  $a$  and  $b$  are components of the vertical dispersion parameter,  $\sigma_z$ ,  $d$  is the grid distance (m),  $r$  is the number of grid blocks that square  $(i,j)$  is from the central receptor square and  $f$  is <sup>a matrix of</sup> 16 point wind direction frequency <sup>divided by windspeed</sup>. The assumptions underlying this equation are outlined by Hanna (1971).

It is assumed for the execution of this model that  $a=.15$  and  $b=.75$  as this approximates neutral conditions and is applicable to the estimation of annual averages (Hanna, 1972). To adequately approach the problem of urban air pollution from both point and area sources a basic Gaussian plume formula for point sources is solved in conjunction with equation (3.2) (Hanna, 1972). The surface concentration of a pollutant emitted by a point source, at strength  $Q_p(\mu\text{gs}^{-1})$ , can be then be calculated using the formula:

$$\chi = \sqrt{2/\pi} \frac{f Q_p}{2\pi \sigma_z^2 r u \frac{H^2}{16}} \exp\left(-\frac{H^2}{2\sigma_z^2}\right) \quad (3.3)$$

where  $H$  is effective source height<sup>(1)</sup> (m),  $r$  is distance of the receptor point from the source (m) and  $f$  is the frequency with which the wind blows towards the sector of interest. The surface concentration due to both area and point sources in a given square is calculated within the computer code for the ATDL model (Hanna, 1972), contained in Appendix A. The computations are based on emissions and meteorological data described below (see section 3.3) and provide arithmetic mean ground level concentrations for the pollutant in a grid format. The ATDL model output selected for this study is as an annual arithmetic mean for each  $4\text{km}^2$  grid square.

### 3.1.2. The Hybrid Extension to the ATDL Model

The generalised hybrid approach can be described by the following steps (Jakeman et al, 1987a).

1. Assume/identify a statistical distribution for the air pollution concentrations over a fixed period, averaging time and location.
2. Develop a deterministic model, from available data, to predict air pollutant concentrations over a reliable range of percentiles.
3. From the results in (2) estimate the parameters of the statistical distribution assumed in (1).
4. Using the parameters derived in (3) construct a statistical distribution of air pollutant concentrations from which the estimates of high pollution levels and their confidence intervals can be obtained.

The primary assumptions of this approach (Jakeman et al, 1987a) are that:

"A1: changes in emissions and meteorology from one period of interest to another (usually from year to year) do not basically affect distributional type at the site of prediction, only the values of the parameters associated with the distribution: and,

(1)  $H \neq$  emission or stack height

A2: the truncated sample distribution generated by the deterministic model output represents order statistics from a random sample of the true distribution of concentrations for the period of interest; the underlying process generating the true distribution should also be stationary"

In this study the hybrid modelling extension of the ATDL model is applied to the Brisbane urban area and is based on the additional assumptions that:

1. the frequency distribution of TSP data can be reliably represented by a two parameter lognormal distribution; and,
2. a simple deterministic relationship between percentiles of pollutant concentration and the inverse of wind speed exists for this urban area.

The relationships between windfield and concentration described by these assumptions have been observed for matched pairs (as opposed to percentiles) in earlier studies (Benarie, 1969, Benarie, 1980) and similar assumptions have been verified using carbon monoxide data for San Francisco, California USA (Knox and Lange, 1974). More recently it has been determined that an inverse relationship exists for matched pairs of a number of pollutants and wind speed, including TSP, in Canberra, Australia (Jakeman et al, 1987b). The validity of the inverse relationship for percentiles in assumption (2) is examined along with all other major assumptions in Chapter 5.

The probability density function,  $f(x)$ , described by the two parameter lognormal distribution of assumption (1) takes the form:

$$f(x) = \frac{1}{x\sigma(2\pi)^{1/2}} \exp\left\{-\frac{\ln(x/a)^2}{2\sigma^2}\right\} \quad (3.4)$$

where  $a$  is the scale parameter or median and  $\sigma$  is the shape parameter for the distribution. This distribution, equation (3.4), is shown to produce acceptable results for describing the distribution of Brisbane TSP data.

Assumption (2) has been defined in the literature (Simpson et al, 1983) as:

$$x_p = K/U_{100-p} \quad (3.5)$$

where  $x(\mu\text{gm}^{-3})$  is the value for the  $p^{\text{th}}$ -percentile of air pollution data,  $K(\mu\text{gm}^{-2}\text{s}^{-1})$  is a constant, and  $U(\text{ms}^{-1})$  is the value corresponding to the  $100-p^{\text{th}}$ -percentile of the wind speed data. This has been established empirically and found to be acceptable for Brisbane data (Simpson et al, 1987). Because of the nature of the lognormal distribution equations (3.4) and (3.5) can be combined such that the shape parameter ( $\sigma$ ) is given by the natural logarithm of the geometric standard deviation  $(\beta)$  of the inverse of wind speed data set.

As shown in Figure 3-1 the emissions based model is linked to the relevant frequency distribution through the scale parameter. For the lognormal distribution the relationship between the arithmetic mean,  $\mu$ , the median,  $\alpha$ , and the shape parameter,  $\sigma$ , is given by:

$$\alpha = \mu \exp\left(-\frac{\sigma^2}{2}\right) \quad (3.6)$$

Where  $\mu$  is estimated by the ATDL model and  $\sigma$  from wind speed statistics<sup>1</sup> (see equation (3.5)),  $\alpha$  is readily calculated using equation (3.6). Given  $\alpha$  and  $\sigma$  the entire frequency distribution follows from equation (3.4). All major assumptions necessary for the valid application of this approach to the Brisbane data are tested in Chapter 5.

### 3.2. THE STUDY AREA

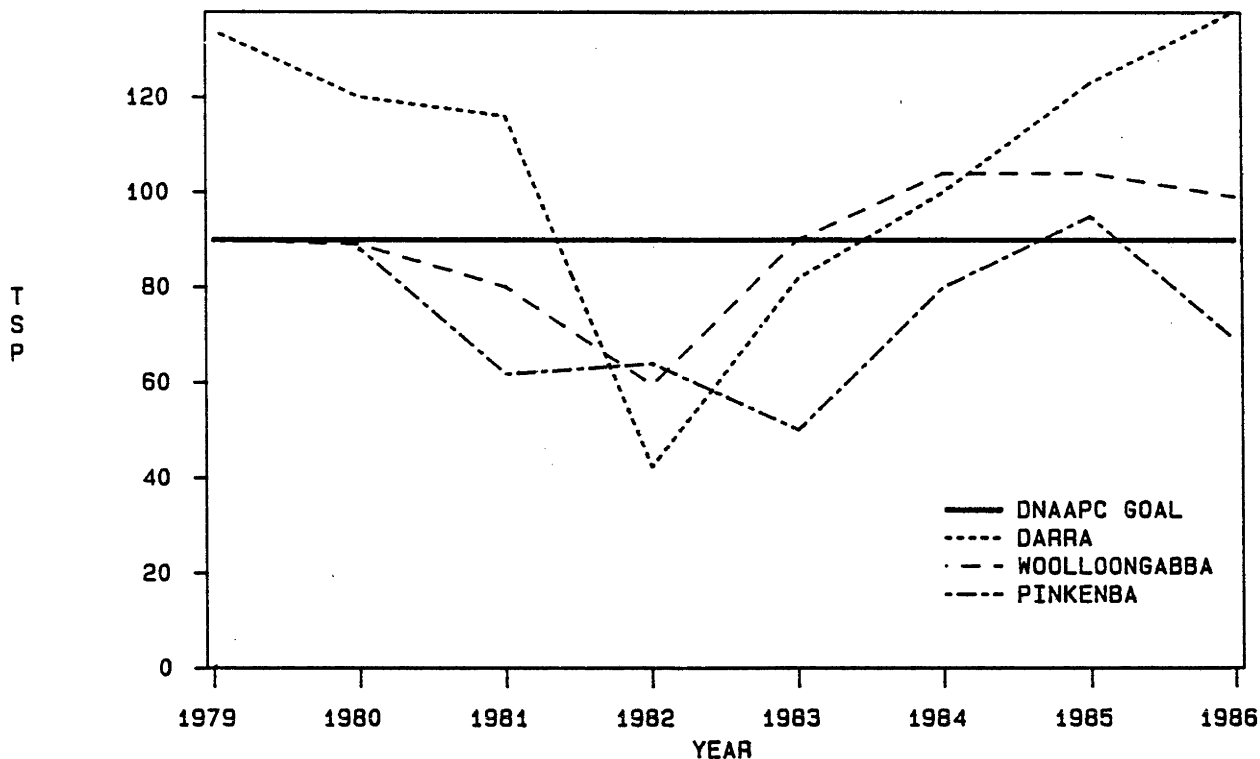
The problems associated with particulate air pollution are of concern in the Brisbane urban area. This is illustrated in Figure 3-2 which shows TSP concentrations in excess of the annual mean ambient air quality goal of  $90\mu\text{gm}^{-3}$  at three sites in Brisbane; Darra (industrial), Woolloongabba (industrial/light commercial) and Pinkenba (industrial/light commercial). The population density in the Brisbane area is greater than 200 persons per square kilometer with a resident population of approximately 1.1 million. Located on the east coast of Australia at 27° 30' S and 153° 00' E it experiences characteristically subtropical climatic conditions. Summer rainfall, with heavy periodic rain, is a dominant feature for this mainly hot and humid climate. The winter is characterised by mild temperatures and some significant rains (Castles, 1986).

Recognising that a problem exists with particulate pollution in Brisbane, and that the population of Brisbane is steadily increasing (Castles, 1986), it follows that the propensity for problems in association with this pollutant will increase. This reinforces the selection of Brisbane as an appropriate study area for the development of the approach outlined above. The data required from the Brisbane airshed for application of the HEC approach, equations (3.2)-(3.6), include emissions and meteorological data. Ambient particulate data are also necessary for model validation. These are discussed in the following section.

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<sup>1</sup>K is not needed for this estimation as it is constant. The geometric standard deviation is therefore derived from the inverse of wind speed

**Figure 3-2:** Annual mean TSP levels,  $\mu\text{gm}^{-3}$ , at Darra, Woolloongabba and Pinkenba sites from 1979-86



### 3.3. THE DATA

The ambient air quality data used for both modelling techniques are 24 hour average TSP values, produced from high-volume sampling techniques, monitored by the Division of Noise Abatement and Air Pollution Control Authority (DNAAPC). Twenty-four hour averaged wind speed data, developed from 3 hourly values monitored by the Bureau of Meteorology in Brisbane have also been collated. Averaging times of 24 hours relate to the guidelines commonly prescribed for TSP and adopted by the DNAAPC. Initially a one year period was used for model development. This was July 1978 to June 1979 which corresponded to the time frame for the emissions inventory that was compiled for the Brisbane area. This was then extended over the years 1979-84 for which additional meteorological data are available.

Examination of the applicability of the approach to other urban areas has required that similar data <sup>excluding emissions data,</sup> have been collated for Sydney, Adelaide, Melbourne and Canberra. To investigate the relationship between these measurements and inhalable and respirable particles additional size selective, nephelometer and microscopic analysis data have been used. The data that have been used for these investigations are detailed in the following subsections.

### 3.3.1. Total Suspended Particulates

The sampling of TSP by high volume procedures is outlined in AS 2724.3 (1984). This stipulates a one day in six monitoring schedule which produces, at best, 60 values per year under uninterrupted monitoring. The only notable variation for the Brisbane data is that the DNAAPC samples at a reducing volume (ie the resistance across the filter due to increased substrate is not compensated in an increased pumping rate). Currently six sites are maintained within the Brisbane air quality monitoring network and these have remained relatively unchanged since 1979. The study area and sampling sites used to collect these data are shown in Figure 3-3.

### 3.3.2. Meteorological Data

The Bureau of Meteorology wind speed data were obtained from the Eagle Farm Airport anemometer. This instrument has limited accuracy at low wind speeds, less than  $0.5 \text{ ms}^{-1}$ , as it is calibrated for the range  $0 - 50 \text{ ms}^{-1}$  and provides approximate 10 minute averages which are collected manually from chart recordings every 3 hours. This will affect the range of the data selected for use with the hybrid modelling approach. However, as the Bureau data from the Brisbane Airport site have been previously used successfully for particulate modelling (Simpson et al, 1986, Simpson et al, 1987), they have, similarly, been selected for use in this study. The Bureau of Meteorology data of this form are also available in many centres throughout Australia and, therefore, the approach proposed in this study may be readily examined in other parts of the country.

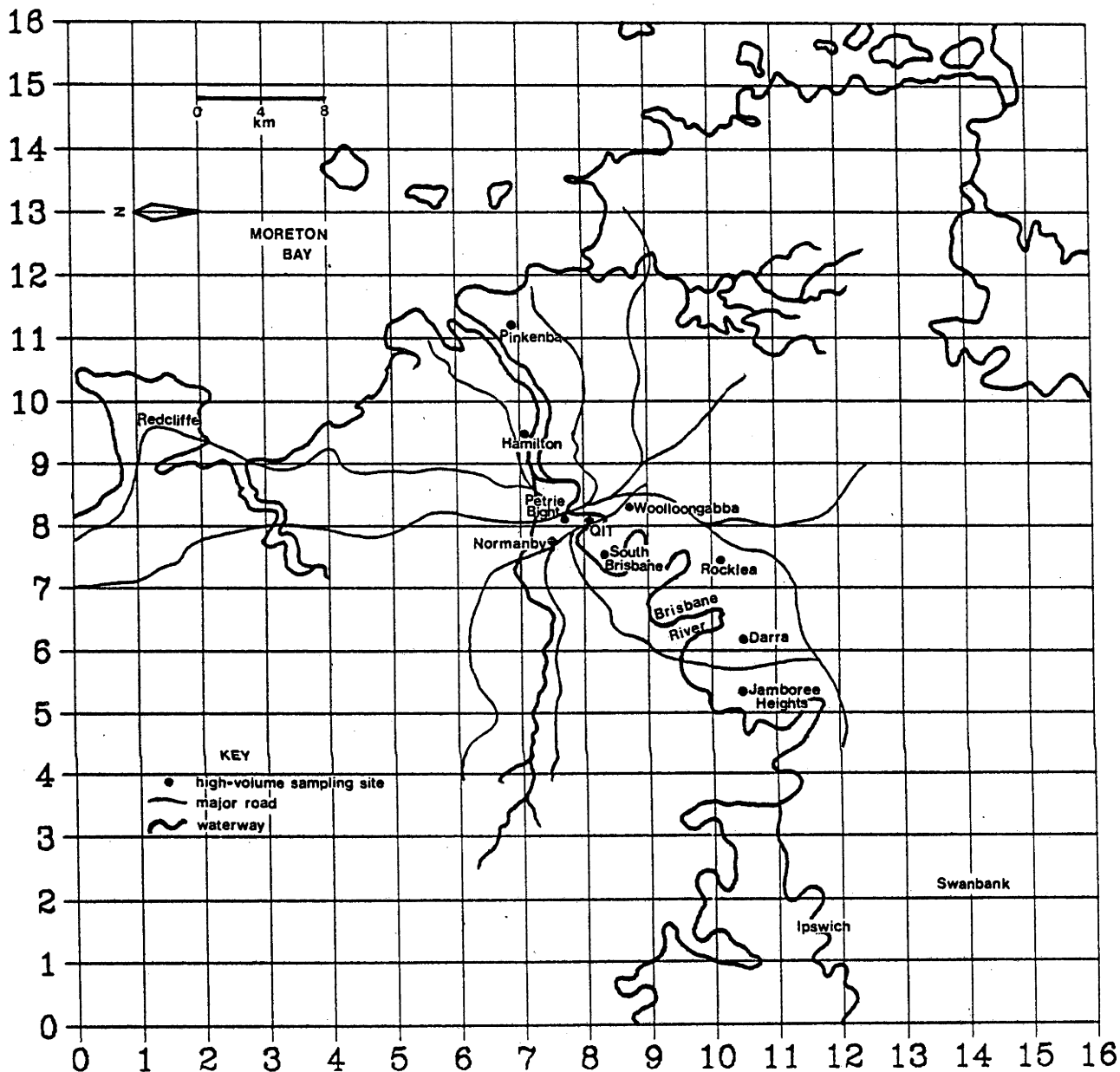
For execution of the ATDL model three hourly Bureau of Meteorology readings of wind direction have been reduced to a yearly average sixteen point wind rose for the Brisbane Airport site. Additional meteorological data such as the components for vertical atmospheric stability,  $\sigma_z$ , have been assumed to be best represented by neutral conditions based on previous experience in the use of this model (Hanna, 1972).

### 3.3.3. Emissions Inventory

The ATDL model also requires emissions inventory<sup>(1)</sup> data. The grid area used for this application of the ATDL model in this study is  $64 \text{ km}^2$  and is divided into 256 squares of  $4 \text{ km}$  by  $4 \text{ km}$ . The DNAAPC emissions inventory is based on irregular grid sizes therefore sources have been aggregated or divided according to their location. The ATDL model places a limit to the number of height classes that may be used in the calculations of plume rise and dispersion. Twenty sources

<sup>(1)</sup> this is complete to the satisfaction of the DNAAPC.

**Figure 3-3:** Map of the TSP monitoring sites within the Brisbane emissions inventory grid network



can be ascribed to each of ten classes. The classes selected were 5, 10, 15, 20, 25, 30, 40, 50, 100 and 150 metres. A total of 106 point sources were allocated to these classes.

The inventory has some limitations which must be outlined. Firstly no background particulate levels have been incorporated and, secondly, fugitive<sup>(1)</sup> emissions have not been accounted for. While it is now possible to estimate a

(1) not accounted for in gross emissions calculations on inventory survey  
eg. losses/emissions through leakage.

plausible background figure for particulates at some sites in Brisbane from chemical composition data (Verrall et al, 1986), fugitive emissions during the 1978-79 period remain unknown.

#### 3.3.4. Chemical Composition

The additional data required to estimate background particulate concentrations have been obtained from a recent TSP composition study conducted by the DNAAPC (Verrall et al, 1986). In this study Verrall et al (1986) use a weighted target transformation factor analysis program, FANTASIA (Hopke et al, 1983), to apportion sources for elements from TSP samples. This form of analysis is based on the assumption that "the amount of a given element is the linear sum of a well defined number of independent sources" (Hopke et al, 1983). The TSP samples were obtained using high-volume samplers with Whatman 41 cellulose filters. The six sites of Darra, Hamilton, Jamboree Heights, Pinkenba, Rocklea and Woolloongabba were monitored during the winter months of July-September, 1985. A total of 372 samples were analysed for 18 elements using X-ray fluorescence techniques.

Crustal TSP, characterised by silica and aluminium, has been primarily attributed to the background sources of re-entrained soil, dust from land clearing and road verges (Verrall et al, 1986). This category of particulates was found to contribute between 40-64% to the average weight of TSP across all sites. This information can be used to produce an average background factor for each of the TSP monitoring sites used in 1985 and a general background concentration that may be used throughout the Brisbane area.

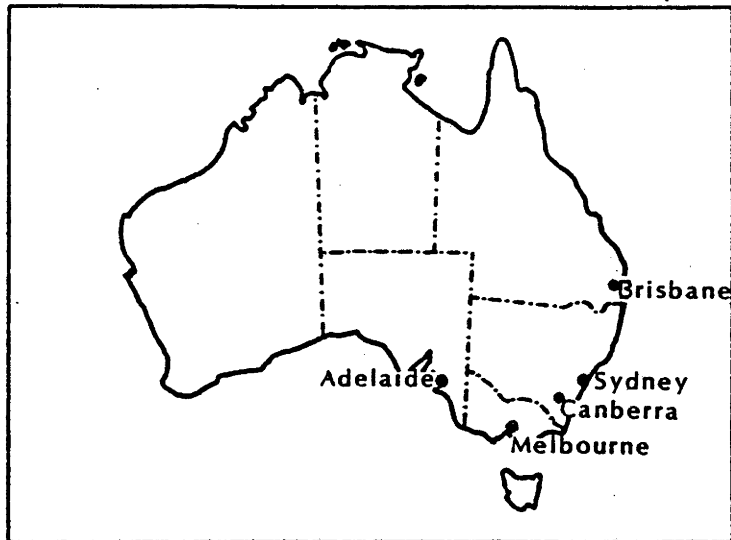
#### 3.3.5. Model Applicability

In order to estimate the potential applicability of the model additional data have been selected from a number of urban areas around Australia. These urban areas represent the major cities of the eastern coast (Brisbane and Sydney), south eastern coast (Melbourne), central southern coast (Adelaide) and south eastern inland (Canberra) regions of Australia. Figure 3-4 shows the location of these urban areas.

The TSP data have been collated from published results of high-volume sampling conducted by the appropriate State or Federal government authorities responsible for air quality. The wind speed data have been made available through the Australian Bureau of Meteorology. The anemometer sites selected for this study are all located at the respective city airports. As mentioned in section 3.3.2, due to the scale and calibration errors for this instrumentation, the low wind speed data are



**Figure 3-4:** Map of urban centres used to test the lognormal and empirical assumptions of the TSP hybrid emissions control model



considered to be suspect. This inaccuracy can be avoided by censoring the wind speed data used for the deterministic component of the hybrid modelling approach (Simpson et al, 1987).

### 3.3.6. Inhalable and Respirable Particles

To examine more directly the relationship between model output and health effects, further size dependent data have been prepared. This has involved the analysis of size selective inlet (SSI) high volume samples, nephelometer data and applying light microscopy, Transmission Microscopy and Scanning Electron Microscopy techniques as well as micro probe analysis to TSP samples.

The SSI high volume sampling procedure used in Brisbane is very similar to that for TSP outlined in AS 2724.3 (1984). The Sierra-Andersen 321A Size Selective Inlets, which replace the normal high volume sampling inlet, operate using principles of impaction. The larger particles are accelerated and impacted allowing particulate matter less than  $10\mu\text{m}$ , commonly referred to as  $\text{PM}_{10}$ , to be collected on a subsequent high-volume filter. The collection efficiency is reportedly independent of wind speeds from 0 to 24 kilometers per hour (Sierra-Andersen, 1980). This is contentious given recent findings in the literature (Wedding et al, 1985). It was found that the 321-A  $10\mu\text{m}$  inlet displayed a tendency to over-sample at high wind speeds. The effects of particle bounce, whereby particles avoid impaction at high wind speeds and become part of the filter mass, can now be mitigated.

$\text{PM}_{10}$  has only been monitored since 1 January 1986. The two sites chosen,

Rocklea and Woolloongabba represent light commercial and light commercial/motor vehicle sources respectively. The monitors run concurrently with TSP high-volume samplers for 24 hours at six day intervals. Additionally a nephelometer is operated continuously at the Rocklea site.

Nephelometers are located at 2 other sites in Brisbane; Fortitude Valley and Eagle Farm. These instruments provide 1/2 hourly averages from 5 minute readings of scattered light,  $b_{\text{scat}}$ , which can then be used to provide estimates of respirable particles. Table 3-1 shows the instruments used at each site and their operating specifications. It can be seen from this Table that the sampling conditions vary between sites. This will affect the scattering light measured by the instrument, particularly for atmospheric conditions with relative humidities greater than 60%. The overall accuracy of each nephelometer is  $\pm 10\%$  of scale, at a wavelength of 530nm and sample flow rate of  $140 \text{ l.min}^{-1}$  (MRI, 1977, MRI, 1980).

**Table 3-1:** Operating specifications for nephelometers in Brisbane

Site	Model	Height of Sample Inlet	Heating of Inlet
Rocklea	MRI 1561	3 m	heated for > 65% RH until 1984
Fortitude Valley	MRI 1591	2 m	heated continuously
Eagle Farm	MRI 1591	3 m	heated for > 65% RH

The microscopic and micro probe analyses of TSP samples were conducted by Dr M. Glikson following procedures developed for the examination of Canberra particulates (Glikson et al, 1987). The analyses were conducted upon TSP samples taken from the 1985 TSP composition study (Verrall et al, 1986). Three days were selected at random from this period for each of the sites.

## Chapter 4

# CONTROLLING FOR THE EFFECTS OF LONG TERM EXPOSURE

### 4.1. INTRODUCTION

Air quality models, used to assist the development and implementation of control strategies, must relate to the standards or goals that are appropriate for the control of the pollutant in question. It would also be an advantage to link these standards to important control variables, such as location and strength of emissions, thereby providing decision makers with information to selectively control sources for the benefit of public health. Benarie (1980) outlines three main requirements for establishing this link:

1. "knowledge of the source strength;
2. adequate definition of the meteorological parameters;
3. reliable methods for the calculation of dispersion from inputs (1) and (2)."

Commonly, ambient air quality standards or goals relate to the frequency distribution for any particular pollutant (Georgopolous and Seinfeld, 1982). For example, a standard may be written in terms of the number of times the maximum, second highest, mean or median concentration is exceeded for a given averaging time. In Brisbane the Queensland Division of Noise Abatement and Air Quality Control (DNAAPC) uses a long term annual arithmetic mean for TSP, advised by the Australian National Health and Medical Research Council (NH&MRC) as a guideline to ambient air quality. The prediction of ground level concentration for this averaging time can be made by a number of models including the Atmospheric Turbulence Diffusion Laboratory (ATDL) model (Hanna, 1972). This model will also relate the "standard-relevant" prediction directly to emissions for the purpose of control decision making.

As outlined in Chapter 2 the composition of particulate air pollution is extremely variable. The sources contributing to TSP concentrations are also varied with the

inclusion of natural as well as anthropogenic emissions (Subcommittee on Airborne Particles, 1979). Difficulties for air quality managers arise in attempting to promote source controls for anthropogenic emissions in terms of a standard for TSP that includes natural emissions. Clearly it is necessary to understand how these respective source categories contribute to any observed TSP level so that only those appropriate sources are controlled. Emissions inventories are used for this purpose but they, like the Brisbane inventory, are commonly compiled only for anthropogenic sources. This leaves any model based on such an emissions inventory struggling to accurately explain variations in ground level concentrations when natural sources contribute significantly.

This Chapter examines how an emission control strategy, based on a long term ambient air quality goal for TSP, can be assisted using a calibrated version of the ATDL model. In this study the ATDL model is run with a particulate emissions inventory of anthropogenic sources in Brisbane. This is augmented by the results of receptor modelling for TSP composition detailed in Chapter 3. With the knowledge of emissions from anthropogenic sources combined with that of background crustal TSP concentrations, an appropriately calibrated model for the prediction of mean annual ground level concentrations can be developed. This is also the first step of the HEC approach, outlined in section 3.1 whereby the deterministic model, providing a basic link between emissions and ground level concentration, can be extended to produce an entire frequency distribution of predicted concentrations.

## 4.2. CALIBRATION OF THE ATDL MODEL

In this section the performance of the ATDL model, described in section 3.1.1, is tested and the subsequent need for calibration is examined. It can be seen from Table 4-1 that the ATDL model based on the 1978/79 emissions inventory tends to underpredict the observed annual average TSP levels by up to a factor of 3.7. This varies between sites and will also vary with the wind speed and frequency data that are used in conjunction with the emissions inventory (Hanna, 1972, Littleboy, 1985). The results from the ATDL model quoted in Table 4-1 are average ground level particulate concentrations corresponding to individual grid squares of 4km<sup>2</sup>. The observed TSP data are annual averages from six high-volume samplers located within five different grid squares (see Figure 3-3).

#### 4.2.1. The Use of Background Crustal TSP

Having noted that the basic emissions inventory does not contain estimates of background particulate levels, an attempt has been made to remove this limitation and thereby improve the ATDL model predictions. This has been done through the use of the recent TSP composition study conducted by the DNAAPC (Verrall et al, 1986). The ATDL model results, based on anthropogenic and predominantly non-crustal sources, are first compared with calculated non-crustal TSP levels. The non-crustal TSP component for each site has been assumed to be a constant proportion of observed TSP and is calculated using the following formula:

$$\chi_{nc} = \chi_o (1-B) \quad (4.1)$$

where  $\chi_{nc}$  is the observed TSP concentration adjusted to exclude background crustal TSP levels ( $\mu\text{gm}^{-3}$ ), B is the proportion of average crustal TSP to the average total TSP at each site for 1985 winter sample<sup>(1)</sup> (dimensionless), and  $\chi_o$  is the observed TSP concentration ( $\mu\text{gm}^{-3}$ ). Of the 1985 sites two were established in 1978/79 and therefore applicable to this period of study. These are Hamilton (B=.63) and Woolloongabba (B=.57).

**Table 4-1:** Comparison of observed and ATDL predicted average annual ground level concentrations of TSP,  $\mu\text{gm}^{-3}$ , including adjustments for background crustal TSP

Site	Observed Arithmetic Mean	Observed Non-crustal Mean	Predicted Arithmetic Mean	Predicted Mean + Crustal
Hamilton	55	16	15	57
Normanby	97	41*	26	68
Petrie Bight	53	23*	34	76
QIT	109	46*	31	73
South Brisbane	58	25*	27	69
W'gabba	99	42	31	73

\*No crustal TSP proportion is directly appropriate as these sites were not monitored during the 1985 study (Verrall et al, 1986).

The factor, B, for Woolloongabba has therefore been used.

The limitations of the emissions inventory can also be circumvented by adding to the ATDL predicted mean concentrations a constant crustal background level. This level has been included as a ground level concentration in the ATDL model attributable to area source emissions (see Appendix A). It is based on the average

*(1) daily variation in background TSP is unknown. Insufficient data is available for this detailed analysis.*

crustal TSP concentration monitored at the Jamboree Heights site during the 1985 TSP composition study (Verrall et al, 1986). This site, of the sites monitored during the composition study, best represents background air quality within the Brisbane urban area due to its location in relation to known particulate sources (Verrall, 1987). The level established for this site during the winter months of 1985 was  $42 \pm 21.1 \mu\text{gm}^{-3}$  from 39 samples. Although not directly applicable to mean levels over an entire year (due to the limited sampling period), it offers the only reasonable and available estimate of background crustal TSP. It has therefore been used for this modelling exercise.

By comparing the ATDL predictions in column 4 with the non-crustal concentrations in column 3 of Table 4-1 it can be seen that the ATDL model estimates of ground level concentrations for anthropogenic particulates are within a factor of two. Column 5 of Table 4-1 shows that the ATDL predictions of TSP (anthropogenic plus crustal background) are also within a factor of 2 if the relatively crude adjustment of  $+42\mu\text{gm}^{-3}$  for background crustal TSP is made. This method of background estimation has been used to calibrate the ATDL model and improve the estimates for all grid squares as opposed to only those squares for which the observed TSP levels have been monitored (from equation (4.1)). In this way the ATDL model is able to predict annual average TSP concentrations that are comprised of both natural and anthropogenic sources.

To use this background level for all squares it must be assumed that the results will not be significantly affected by the temporal and geographical variation in emissions from crustal sources. As there is considerable daily variation in the crustal background estimate as demonstrated by the standard deviation ( $42 \pm (21)$ ) on the Jamboree Heights crustal estimate (Verrall et al, 1986), this uncertainty must be acknowledged in assessing model output. Variation in wind speed and frequency is also known to affect estimates of pollutant concentration using the ATDL model. An improvement in this modelling technique may be to incorporate more wind field data over the grids under investigation.

It should also be noted that the use of crustal TSP as a background may overlap in some areas with the emission inventory where estimation of source strengths from industries, such as quarrying, will include crustal particulate matter. This may falsely elevate predictions in some areas but will not affect the overall spatial estimates as the background calibration ( $+42$ ) has been derived from a site well removed from the dominant particulate sources.

### 4.3. CONTROLLING FOR THE MEAN

Figure 4-1 shows the spatial distribution of the ATDL model estimates using a UNIRAS contour plotting routine, Geopak routine GCONR2. Using the crustal background estimate of  $42\mu\text{gm}^{-3}$  to adjust the ATDL model output the mean goal of  $90\mu\text{gm}^{-3}$  would be exceeded in 5 grid squares corresponding to Petrie ( $103\mu\text{gm}^{-3}$ ), Lytton ( $108\mu\text{gm}^{-3}$ ), Pinkenba ( $93\mu\text{gm}^{-3}$ ), M<sup>t</sup> Cootha ( $91\mu\text{gm}^{-3}$ ) and Holland Park ( $134\mu\text{gm}^{-3}$ ). These hot spots are characterised by differing combinations of point and area sources.

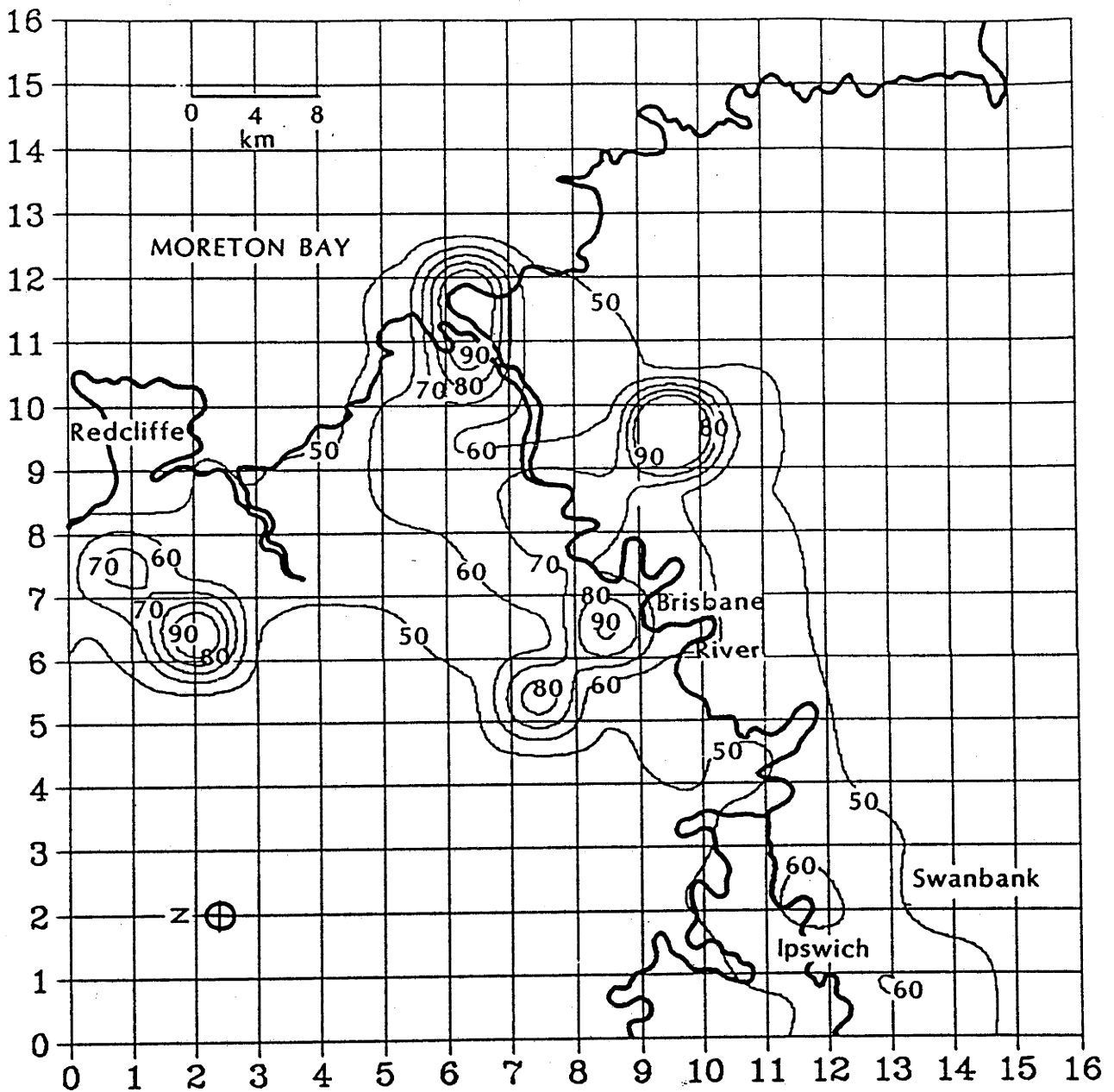
Table 4-2 demonstrates the range of source configurations for the monitored grid squares. It can be seen that within the category of area sources the car is the dominant emitter by weight. These sources, however, may not completely dominate the predicted ground level concentrations for their respective grid squares. It is more appropriate to consider all sources from surrounding grid squares that contribute to the prediction of ground level concentrations. This is shown in Table 4-3 for the sites where the DNAAPC goal is exceeded.

Table 4-3 contains the ratio of point to area source contributions to the ATDL predictions. As the area source contribution increases the ratio approaches one. The information contained in Table 4-3 can be used to identify whether an emissions control strategy should be directed towards point and or area sources in the proximity of the grid square. For the purposes of emission control the non-crustal ratios are the most useful. This ratio refers only to the controllable anthropogenic emissions thereby excluding estimated background crustal sources.

The sites of Petrie, Holland Park and M<sup>t</sup> Cootha are hot spots dominated by point sources with effective stack heights of 5m. These dominant sources have been identified as quarries in the Brisbane emissions inventory (Borowski, 1984). The effects of source specific emission reduction on ground level concentration can then be determined. The control of area sources would be most appropriate for the sites of Lytton and Pinkenba and, as area source emissions are dominated by motor vehicles, the extent to which the DNAAPC can regulate vehicle emissions must be outlined.

If it is assumed that gross particulate emissions from all cars and equivalent vehicles will be reduced by approximately 24% by 1990 through the introduction of unleaded petrol (Stewart et al, 1983), then given estimated car emissions the reduction in ground level TSP concentration can be estimated. The results of the model execution under these conditions demonstrate that, while the Pinkenba site becomes acceptable, the Lytton site would remain in violation of the DNAAPC

Figure 4-1: Contour plot using UNIRAS Geopak routine GCONR2 of ATDL annual mean estimates for TSP,  $\mu\text{gm}^{-3}$ , adjusted for crustal background





**Table 4-2:** Emission rates for dominant sources  
(in tonnes per annum) within the monitored grid squares

Source	Character	Hamilton	Petrie Bight	Normanby	South Brisbane	Woolloongabba QIT
Area	Cars	38.729	158.670	106.872	96.850	192.330
	Trucks	4.906	45.950	16.161	18.305	9.854
	Domestic	8.301	14.000	13.525	14.017	16.441
	Total	51.936	219.530	136.558	129.175	218.625
Point	Dominant	4.807	222.000 109.300	249.800	154.000	3.750
	Total	4.817	343.200	271.213	154.065	4.225

**Table 4-3:** Ratios of area to point source contributions to ATDL predicted ground level concentrations for grid squares where the DNAAPC goal for annual mean TSP is exceeded

Site	Area to Point Ratio Non-crustal	Area to Point Ratio Total
Petrie	.0982	.4669
Holland Park	.1997	.5476
M <sup>t</sup> Cootha	.2045	.5727
Pinkenba	.9018	.9399
Lytton	.8309	.9090

goal of  $90\mu\text{gm}^{-3}$ . Nonetheless the reduction achieved in the predicted ground level TSP concentration would be from 108 to  $94\mu\text{gm}^{-3}$  which is well within the accuracy of model predictions. At other sites, where area source emissions do not dominate and where reductions in ground level concentrations of greater than 15% are required, additional control strategies should be examined.

#### 4.4. APPLICABILITY TO CURRENT STRATEGIES

To examine the applicability of the ATDL model based on 1978/79 emissions to current management strategies in Brisbane, the model has been executed using the 1978/79 emissions inventory and annual wind speed and direction data between 1979 and 1984. Table 4-4 demonstrates that the ATDL model provides predictions of the annual arithmetic mean, within a factor of 2, at most sites for most years. The notable exception to this is the Darra site. The under-estimation of TSP concentrations by the ATDL model at the Darra site can be largely explained by

fugitive emissions from a large cement factory in the proximity of the monitor. These fugitive emissions are not included in the emissions inventory and therefore not accounted for in the model predictions.

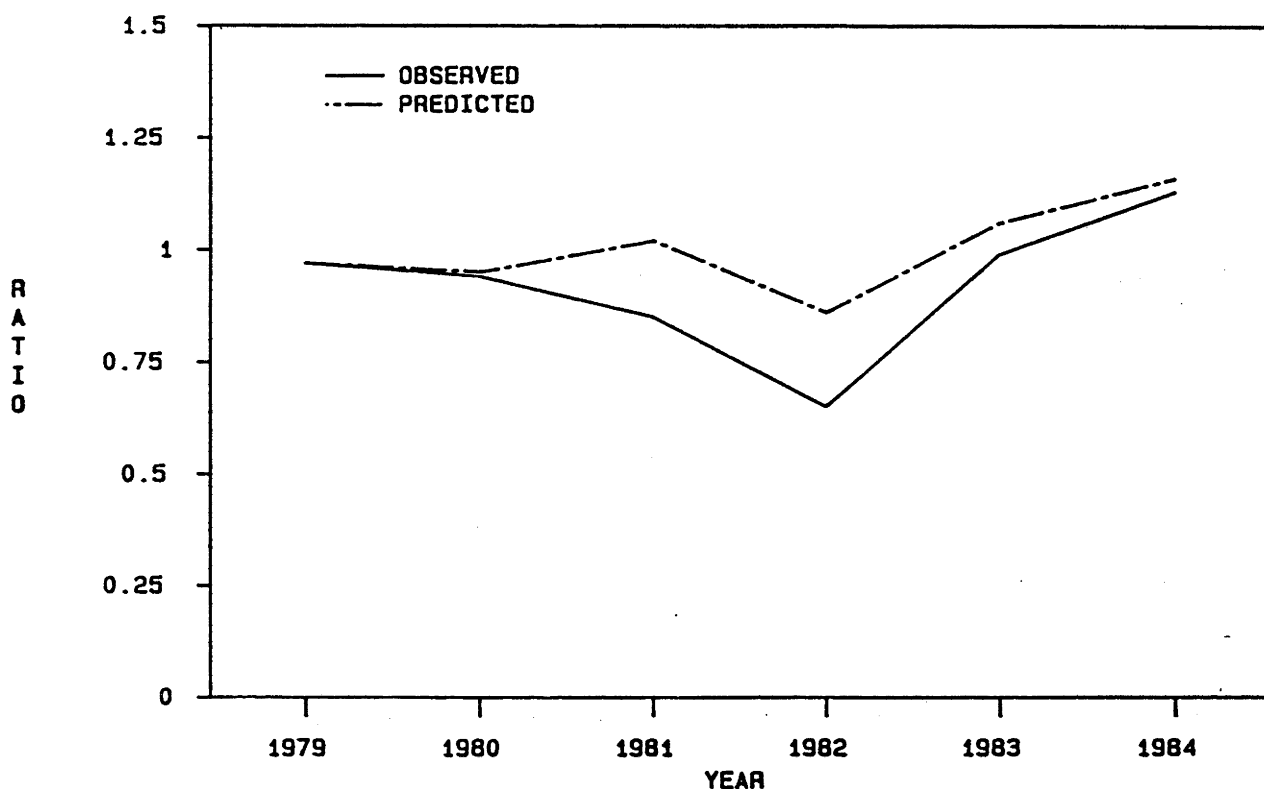
**Table 4-4:** Observed non-crystal\*  $\chi_o$  and ATDL predicted non-crystal  $\chi_p$  mean ground level concentrations of TSP,  $\mu\text{gm}^{-3}$

Year	Variable	Hamilton	W'gabba	Darra	Jamboree	Pinkenba	Rocklea
1979	$\chi_o$	22	41	86	16	-	29
	$\chi_p$	14	30	16	15	-	15
1980	$\chi_o$	23	40	68	15	41	27
	$\chi_p$	14	29	15	14	13	14
1981	$\chi_o$	19	36	67	13	31	26
	$\chi_p$	20	41	21	21	17	19
1982	$\chi_o$	-	27	23	13	29	40
	$\chi_p$	-	38	19	19	16	17
1983	$\chi_o$	18	42	46	10	24	29
	$\chi_p$	17	36	19	19	15	17
1984	$\chi_o$	20	48	57	13	39	34
	$\chi_p$	17	37	20	17	17	18

\*calculated using equation (4.1)

Figure 4-2 also shows that, for the Woolloongabba site, the assumption that emissions have remained relatively constant is quite acceptable. This Figure demonstrates that concentrations of TSP predicted by the ATDL model closely mirror the observed concentrations for most years. It should be noted that although total emissions are seen to remain constant the relative contributions of point and area sources may have altered. Area source emissions may have increased while controls upon point sources may have meant that total emissions appear relatively unchanged. The ATDL model, however, deals with area and point source dispersion in different ways allowing any marked change in the relative contributions to ground level concentrations to be detected.

**Figure 4-2:** The ratio of predicted ATDL (1979-84) to predicted ATDL (1978/79) compared with the ratio of observed (1979-84) to observed (1978/79) mean TSP concentrations for Woolloongabba



#### 4.5. DISCUSSION

Currently standards for TSP are cited in terms of total mass and therefore include crustal material. The basic emissions inventory of anthropogenic sources used in this study, however, does not include crustal emissions that are generated from natural processes. These background crustal particulates are generally regarded as less hazardous to human health due to their size and composition. Nonetheless, information on the contribution of crustal material to ambient TSP is necessary to produce models that relate to TSP goals currently used by the DNAAPC.

The ATDL model has been applied to the Brisbane urban area and shown to estimate ground level non-crustal particulate concentrations to within a factor of 2 of calculated non-crustal TSP. This level of accuracy has been found to be the best achievable for modelling air pollutant concentrations (Hanna, 1982) and is largely irreducible due to the stochastic nature of atmospheric processes (Hayes and Moore, 1986). When combined with an estimate of background crustal concentration, derived from analyses of chemical composition, the ATDL model can also be shown to provide acceptable (crustal + non-crustal) estimates of annual mean ground level TSP concentrations. The assumptions made when incorporating estimated background crustal levels restrict the extent to which these results can be validated and applied. Nevertheless this calibrated ATDL model provides a

useful tool for the spatial analysis of TSP concentrations and the identification of "hot spots" or high concentration areas. It has also been shown that emissions have not changed substantially over the 1978/79-84 period and therefore any conclusions relating to emissions control are probably quite relevant to present air quality control in Brisbane.

The output from the ATDL model also enables the assessment of relative contributions of point and area sources to the resultant ground level concentrations. This is shown to be a very useful guide to the development of an appropriate emissions control strategy for TSP in Brisbane. The control of point sources can be approached on an industry by industry basis through the normal licensing and review procedures available to the DNAAPC under the regulations of the Clean Air Act (1962-1986). The grid squares that have been identified as requiring point source controls include Petrie, Holland Park and M<sup>t</sup> Cootha. The Lytton and Pinkenba sites require predominantly area source controls. These area sources, comprising car, truck and domestic emissions, are largely beyond the jurisdiction of the DNAAPC. The introduction of unleaded petrol, however, may be sufficient given current vehicle volumes, to reduce the particulate emissions from cars, and thereby the ground level TSP concentration, to within acceptable limits.

The question must be asked, is it sufficient to manage urban air quality in terms of long term annual averages alone? It is known that acute effects to human health can become manifest at averaging times of 24 hours and less (Stern et al, 1984) and that information on short duration high concentration exposures will be lost using data averaged over longer periods. The problem of developing a model that will estimate ground level concentrations relating to a number of goals, primarily related to short term exposure, is addressed in the following Chapter.

## Chapter 5

# CONTROLLING FOR THE EFFECTS OF SHORT TERM EXPOSURE

### 5.1. INTRODUCTION

The goals used to ensure that ambient air quality is suitable for the maintenance of community health are often a combination of short and long term standards. Indeed, it has been shown that controlling emissions in terms of long term goals may not ensure that short term exposure remains within prescribed limits (Simpson et al, 1986). As outlined earlier, air quality models to be used for the development and implementation of control strategies must relate to these standards or goals that are appropriate for the pollutant to be controlled. However, many different standards may be applied to a pollutant at any one time, and given that standards change over time, it would clearly be beneficial for model output to be flexible.

In Chapter 4 it was stated that the long term annual arithmetic mean for TSP of  $90\mu\text{gm}^{-3}$ , used by the Queensland DNAAPC, provides a guideline to ambient air quality in Brisbane. It was also demonstrated that long term exposure to particulate concentrations may be assessed in terms of this annual average using the ATDL model calibrated through the use of chemical composition data.

To adequately protect human health from the effects of air pollution, however, it is necessary to control short duration high concentration exposure as well as long term exposures of lower concentration. These short term exposures are described by the upper percentiles of the frequency distribution for a given pollutant. To implement emissions controls which avoid violations of other goals, such as the USEPA 24 hour maximum standard of  $260\mu\text{gm}^{-3}$  for TSP, a new model must be developed or models such as the ATDL model must be extended.

This Chapter examines how an emissions control strategy, using more than one air quality goal, can be developed based upon the results given by the calibrated ATDL model in the previous Chapter. It is known that deterministic models, such as the ATDL model, do not predict extreme events well (Jakeman et al, 1987a).

The ATDL model alone is also limited to estimating annual average concentrations in Brisbane as the emissions data, used to make these predictions, are only available as annual averages. Using assumptions about the statistical nature of air pollutant concentrations and a deterministic relationship between wind speed and particulate air pollution, the hybrid modelling approach is used here to relate this annual average concentration to other relevant air quality criteria. The final TSP hybrid emissions control (HEC) model that is developed can predict the entire frequency distribution of particulates using emissions and meteorological data, thereby providing estimates of any useful statistic of that distribution, such as the 24 hour maximum concentration. These ambient concentrations can then be linked through the ATDL model to specific emissions thereby allowing appropriate control strategies to be developed.

## 5.2. THE HYBRID APPROACH

The hybrid modelling approach has been outlined in Chapter 3 with a full description of the approach found in a series of papers by Jakeman, Simpson and Taylor (1986, 1987a,b). Essentially this approach combines both statistical and deterministic or empirical modelling techniques to predict concentrations of air pollutants; hence the term hybrid. The following subsections describe how the approach has been applied to this study and provide justification for its use in extending the ATDL model.

Table 1-1 contains the variety of source conditions to which the hybrid approach has been applied to date. In one of these applications to the problem of urban particulate pollution in Brisbane, Australia (Simpson et al, 1986, Simpson et al, 1987), a simple, almost analytic, approach has been used. This has involved the assumptions, detailed earlier in section 3.1.2:

1. that the lognormality of the TSP data is consistent for all data sets; and,
2. the simple inverse relationship between the percentiles pollutant concentration and wind speed is consistent for those percentiles used to generate the model distribution.

If this model is to provide the basis of the hybrid modelling for the Brisbane 1978-79 data under investigation then these assumptions, and the general assumptions A1 and A2 outlined in section 3.1.2, must be satisfied, *as they have been in previous studies (eg. see Simpson et al 1986, Simpson et al 1987).*

### 5.2.1. The Identification of the Statistical Distribution

The first step of the hybrid modelling approach is to identify the most appropriate distribution for the pollutant under investigation, in this case TSP. This requires that the data are independently and identically distributed as outlined in assumption A2 (see section 3.1.2.) Given that TSP samples are taken once every six days it is believed that one measurement is largely independent of the next and not likely to be autocorrelated. The question of identical distribution or stationarity has been examined graphically and the results are shown in Appendix B. From these figures it has been assumed that seasonal variation leading to non-stationarity is not sufficiently pronounced to invalidate this technique. It is noted, however, that some extreme events exist within these data sets that may reflect errors in monitoring or single episode large scale emissions (eg Normanby and QIT).

Using a combination of maximum likelihood techniques and the Kolmogorov Smirnov test for goodness of fit the most appropriate distribution is selected from the two parameter lognormal, gamma, Weibull, normal and one parameter exponential distributions (Taylor, 1985). Taylor (1985), based on extensive simulation, outlines a stringent selection procedure for these distributions when using the above tests in combination:

1. the exponential distribution is accepted if the Kolmogorov ratio calculated for this distribution is the lowest,
2. else the maximum value of the log-likelihood functions identifies the most acceptable distribution;
3. a distributional form is only accepted if the corresponding Kolmogorov ratio is less than the confidence level cutpoint (ie less than one for the 95% confidence level).

The Kolmogorov ratio mentioned above is given by:

$$T = \frac{\sup_x |F(x) - S(x)|}{D_{95}} \quad (5.1)$$

where  $F(x)$  is the hypothetical distribution function for  $x$ ,  $S(x)$  is the empirical distribution function for  $x$  and  $D_{95}$  is the 95% confidence level for the Kolmogorov statistic. If this statistic is less than one then the null hypothesis is accepted ( $p < 0.05$ ); that is, the empirical distribution function based on a random sample is not significantly different at the 95% confidence interval from the hypothetical distribution (Conover, 1980). Based on simulations Taylor (1985) showed that the log-likelihood function provided a more powerful method of model selection than the Kolmogorov statistic particularly at small sample sizes (ie approximately 50). Hence the selection hierarchy outlined above is used.

**Table 5-1:** Distributions identified using the maximum of the log-likelihood function and the corresponding Kolmogorov ratio for the lognormal distribution

Site	Number of Samples	Maximum Likelihood Preferred Distribution	Kolmogorov Ratio for Lognormal Distribution
Hamilton	50	gamma*	0.617
Normanby	34	lognormal	1.235
Petrie Bight	49	lognormal	0.683
QIT	34	lognormal	0.625
South Brisbane	27	lognormal	0.612
W'gabba	58	lognormal	0.696

\* the lognormal distribution is also acceptable using the Kolmogorov ratio

The results of this analysis can be seen in Table 5-1. Of the six TSP data sets all except Hamilton are accepted as lognormal distributions using the maximum of the log-likelihood function as the selection criterion. Using the Kolmogorov ratio, equation (5.1), Hamilton can be accepted as lognormal though the Normanby site is not. These tests in combination indicate that the two parameter lognormal distribution is most appropriate to represent the six sites under investigation. This selection supports previous research that has shown the two parameter lognormal distribution to be appropriate for annual TSP data sets at similar monitoring sites in Brisbane between 1979 and 1984 (Simpson et al, 1987). While this reinforces the validity of assumption A1 (see section 3.1.2) it is noted that this may not always be the case and, given the assumptions of statistical distribution models, it is important to identify the appropriate distribution for each new data set.

The probability density function for the pollutant concentration,  $\chi$ , from a two parameter lognormal distribution is given by equation (3.4). The parameter estimates from fitting this function, using maximum likelihood techniques, to the six TSP data sets are shown in Table 5-2. This illustrates the variation between sites in the parameters for this distributional form.

These parameters can then be used to estimate values of the probability density function, equation (3.4). Larsen (1969) found that percentile values of pollutant,  $\chi_p$ , could be determined for a given two parameter lognormal distribution, used to represent the range of values for that pollutant, by calculating:

$$\chi_p = \alpha_{\chi}(\beta_{\chi})^z p \quad (5.2)$$



**Table 5-2:** Median,  $\alpha_\chi$ , and geometric standard deviation,  $\beta_\chi$ , for two parameter lognormal distributions of raw TSP,  $\mu\text{gm}^{-3}$ , data 1978-79

Site	$\alpha_\chi$	$\beta_\chi$
Hamilton	51.6	1.50
Normanby	85.0	1.51
Petrie Bight	50.1	1.39
QIT	84.8	1.73
South Brisbane	50.7	1.35
W'gabba	92.6	1.43

where  $\beta_\chi$  is the standard geometric deviation for  $\chi$ ,  $\alpha_\chi$  is the geometric mean or median and  $Z_p$  is the number of standard deviations from the mean corresponding to the  $\chi_p$  being calculated.

### 5.2.2. The Empirical Relationship

Simpson et al (1986) demonstrated that a simple hybrid model was appropriate for TSP data, and to a lesser extent light scatter data (Simpson et al, 1987), in this urban area. The data in these studies were found to be best represented by a two parameter lognormal distribution and the deterministic model for describing the empirical relationship between wind speed and TSP concentration was of the following form, described earlier in equation (3.5):

$$\chi_p = \frac{K}{U_{100-p}}$$

K from this equation has been calculated using the median pollutant concentration,  $\alpha_\chi$ , and median wind speed,  $\alpha_u$ , as shown in equation (5.3).

$$K = \alpha_\chi \alpha_u \quad (5.3)$$

The validity of this relationship was examined for 1978/79 data. If the relationship is appropriate K will remain relatively constant across a range of percentiles. It was found that this was generally true between the 10<sup>th</sup> and 50<sup>th</sup> percentiles (see Appendix C). The relationship was weakest at the QIT, South Brisbane and Normanby sites. This may be attributed, at least in part, to the incompleteness of the annual data set for these sites. As outlined in Chapter 3 low wind speeds could contribute to the relationship not being consistent for percentiles greater than 50. The validity of this relationship is, however, less

important than its implications for the computing the geometric standard deviation for air pollutant concentrations. In providing an estimate of the geometric standard deviation wind speed is used as another measure of the variation for TSP.

### 5.2.3. The Hybrid Model

In linking the output from this deterministic model in equation (3.5) to Larsen's formula for the calculation of a lognormal distribution, equation (5.2), the following equation was formed:

$$\chi_p = \frac{K}{\alpha_u} (\beta_u)^{z_p} = \alpha_\chi (\beta_u)^{z_p} \quad (5.4)$$

where  $\beta_u$  is the standard geometric deviation for the inverse of wind speed. In order to predict pollutant concentrations for the percentiles of interest the parameters for the assumed statistical distribution must be estimated from the output of the deterministic model. To derive estimates for the scale and shape parameters for this equation from a censored distribution, maximum likelihood techniques have been found to be most appropriate (Jakeman et al, 1987a). Using algorithms developed for this purpose (Taylor, 1985), a lognormal distribution has been fitted to the  $\chi_p$  values between  $\chi_{10}$  and  $\chi_{50}$  from equation (3.5).

**Table 5-3:** Median,  $\alpha_\chi$ , and geometric standard deviation,  $\beta_u$ , for two parameter lognormal distributions of censored output from the deterministic model described in equation (3.5)

Site	$\alpha_\chi$	$\beta_u$
Hamilton	52.8	1.61
Normanby	77.8	1.61
Petrie Bight	51.3	1.61
QIT	85.7	1.61
South Brisbane	50.8	1.61
W'gabba	94.7	1.61

Since it is based on the wind speed data from one Bureau of Meteorology during 1978/79,  $\beta_u$  is a constant as shown in Table 5-3. This value for  $\beta_u$ , 1.61, is representative<sup>(1)</sup> of the  $\beta_\chi$  values in Table 5-2 which are of the range 1.35-1.73. Through this empirical relationship the wind speed data provides a reasonable estimate of the variation for the predicted pollutant concentrations. The exponentiated scale and shape parameters estimated from equation (3.5), providing estimates of the median,  $\alpha_\chi$ , and geometric standard deviation,  $\beta_\chi$ , respectively,

<sup>(1)</sup> other factors will influence  $\beta_\chi$  and it has been found that not all the variability in the TSP data can be explained by the windspeed data alone (Simpson et al 1987).

can then be used in equation (5.4) to predict the TSP concentration for the percentile of interest. This method does not, however, provide any measure of the uncertainty associated with predicting these concentrations.

It is possible to provide some measure of uncertainty by estimating the confidence intervals for selected percentiles. These confidence intervals may be approximated through the use of Monte Carlo simulation techniques. This has been possible for the 1978-79 data under investigation by using an algorithm provided by Taylor (1986). The technique uses the parameter variance-covariance matrix derived during the maximum likelihood estimation. Pairs of scale and shape parameters are selected at random from this distribution for each of the 200 Monte Carlo simulations. These simulations then provide an estimate of the 95% confidence interval for the percentile of interest. Jakeman and Taylor (1985) use this approach for obtaining confidence limits on the 98<sup>th</sup> percentile prediction of 24 hour acid gas levels. They note that these are lower bounds on the intervals because the assumption, that the data are independently and identically distributed, used by the maximum likelihood algorithm may be violated in practice.

Table 5-4 compares the hybrid model estimated 24-hour maximum concentrations of TSP, generated using maximum likelihood and Monte Carlo techniques, with the observed and expected maximum values for TSP. The observed maximum values are extracted from the raw data whereas the expected values are calculated from equation (5.5) using Larsen's model (1969) for inferring concentrations from lognormal data:

$$\chi_{\max} = \alpha_{\chi}(\beta_{\chi})^{2.94} \quad (5.5)$$

If the data are independently and identically distributed the expected 24-hour maximum concentration for a 365 day year (from equation (5.5)) provides a more accurate estimate than the observed maximum that is derived from a yearly sample of one day in six. The parameters for this calculation are those shown in Table 5-2. The value for Z of 2.94 standard geometric deviations from the median is shown by Larsen (1969) to be appropriate for 24-hour averaged data sampled over the period of a year (ie N=365). There is reasonably good agreement, within a factor of 2, between the predicted TSP maxima and the observed and expected TSP maxima. The expected maximum lies within the 95% confidence interval of the hybrid model prediction,  $\chi_p$ , for four of the six sites (ie QIT, Hamilton, Normanby and Woolloongabba).

The model has only been validated over a limited period of one year and is providing annual estimates hence Table 5-4 only shows the spatial variation in

model performance. However this can be supported temporally by the research of Simpson et al (1986a, 1986b) who found a similarly acceptable fit between this model and observed and expected TSP levels for Brisbane over 5 years of data from 1979-1984.

**Table 5-4:** Hybrid model predicted maximum concentrations, -95% confidence interval ( $\chi_P$ ) +95% confidence interval, compared with observed,  $\chi_O$ , and expected,  $\chi_E$ , maxima

Maximum ( $\mu\text{gm}^{-3}$ )	Hamilton	Normanby	Petrie Bight	QIT	South Brisbane	W'gabba
$\chi_O$	113.0	559.0	92.0	429.0	98.0	203.0
$\chi_E$	170.0	285.5	131.9	424.9	122.5	265.0
$\chi_P$	146(197)270	215(289)397	142(191)262	237(319)438	140(189)260	261(352)484

In earlier studies this hybrid model has been used to assist control strategy development <sup>(eq see Simpson et al 1987)</sup>. By simplifying equation (5.4), or rearranging equation (5.5), the following equation can be developed:

$$\frac{\chi_{\max}}{\alpha} = \beta^{2.94} \quad (5.6)$$

This equation determines a value for  $\beta$  that, if exceeded, the maximum standard may be violated whilst the median may remain within acceptable limits and *vice versa*. The long term ambient air quality goal adopted by the DNAAPC is an annual arithmetic mean which may be combined with the USEPA annual 24-hour maximum of  $260\mu\text{gm}^{-3}$  by relating the mean,  $\mu$ , to the median,  $\alpha$ , through the standard deviation,  $\beta$ . Assuming a lognormal distribution equation (5.6) becomes:

$$\beta^{2.94} = \frac{\chi_{\max}}{\mu / \exp(.5 \ln^2 \beta)}$$

A numerical solution to this non-linear equation, in terms of  $\beta$ , gives a value for  $\beta$  of 1.47.

This value is exceeded by three<sup>1</sup> of the six data sets in Table 5-2. This indicates that the short term maximum standard may be violated if emission control strategies were based solely on the the long term annual arithmetic mean standard at the sites of Hamilton, Normanby and QIT during 1978-79. To maintain the integrity of both standards it is necessary to regulate emissions based in terms of both annual average and maximum concentrations. It is noted that

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<sup>1</sup>the site of Normanby is included in this total although it was not accepted as lognormal by the Kolmogorov ratio

conditions will exist where the  $\beta$  value of 1.47 will be exceeded even though neither the maximum nor mean standards are actually violated. This can be seen at the Hamilton site, where although  $\beta_{\chi}$  is greater than 1.47, both the median (see Table 5-2) and the maximum levels (see Table 5-4) fail to exceed the set standards. However, if emissions were allowed to increase producing mean TSP levels just within the standard then this method simply suggests that the distribution of TSP values at that site may have sufficient variation, represented by  $\beta_{\chi}$ , to produce unacceptably high maximum concentrations.

Another method has been proposed in the literature that relates hybrid model output to total emissions (Simpson and Jakeman, 1984, Simpson et al, 1986). This rollback technique uses equation (5.4) directly to produce a possible emission control strategy for avoiding violations of the maximum standard. Given that the maximum standard of  $260\mu\text{gm}^{-3}$  is not to be exceeded, and assuming K depends only on emissions, the equation can be calculated for each site and the percentage reduction required to balance the equation can be estimated. From this method it would be recommended that emissions be reduced at Normanby ( $\approx 25\%$ ), QIT ( $\approx 24\%$ ) and Woolloongabba ( $\approx 31\%$ ).

In relating the hybrid model to emissions the methods presented here provide useful information on potential source controls where such emissions can be treated as an aggregated single source. However, in linking both mean and maximum predictions to an emission control strategy, it would be beneficial to provide source specific information whenever possible. Sufficiently detailed information is required for regulating authorities to make effective decisions on what sources should be controlled and by how much. It has been demonstrated in Chapter 4 that for the same time period, 1978/79, detailed links between annual average concentrations and emissions can be made through the use of models such as the ATDL model.

### 5.3. THE HYBRID EMISSIONS CONTROL MODEL

To improve the link between emission control strategies for both short and long term standards the hybrid model developed in this Chapter can be combined with the ATDL model that has been validated and calibrated in Chapter 4. Having reasonably satisfied the individual assumptions for both the hybrid and ATDL models then linking the outputs of each model is relatively straight-forward. This section will outline this link.

The ATDL model output is an average surface concentration for each 4km by 4km grid square. This may be related directly to the DNAAPC annual mean

guideline of  $90\mu\text{gm}^{-3}$ . To relate this to ambient air quality goals in terms of median and maximum concentrations the following equation may be used:

$$\mu_{i,j} = \alpha_{i,j} \cdot \exp(.5 \ln^2 \beta_u) \quad (5.7)$$

In this way the median,  $\alpha$ , can be calculated from the mean concentrations estimated for each grid square (i,j) by the ATDL model. The fundamental assumptions that must be met here are that, (1) a lognormal distribution is valid throughout the grid network used for the ATDL model, and that, (2)  $\beta_u$  is an appropriate estimate for the geometric standard deviation of this distribution. So far this has only been validated for the grid squares corresponding to the TSP monitoring sites in Brisbane between 1978 and 1984. It is uncertain how different geographical and meteorological conditions within the Brisbane region would affect the distribution of TSP data.

With these assumptions noted for the ATDL model the annual maximum 24-hour concentration,  $\chi_{\text{max}}$ , can be calculated for lognormally distributed data using a version of equation (5.5) that is shown in equation (5.8).

$$\chi_{\text{max}(i,j)} = \alpha_{(i,j)} \beta_u^{2.94} \quad (5.8)$$

The ATDL model is executed using the emissions inventory and wind speed data described in section 3.3. The median and maximum levels for each grid square are calculated using equations (5.7) and (5.8). These equations have been incorporated in the computational code detailed in Appendix A.

To ensure that the HEC model output is realistic the background level used to calibrate the ATDL model (see Chapter 4) must be applied. This level is included as a ground level concentration attributable to area source emissions and is based on the Jamboree Heights site background level detailed in section 4.2.1. The problems in attempting to extrapolate this background level to all squares have also been outlined in the previous Chapter.

Table 5-5 shows that, using a set background figure of  $42\mu\text{gm}^{-3}$ , both mean and maximum predictions are accurate to approximately within a factor of 2 for those grid squares that have been monitored for TSP during 1978-79. The expected maximum is calculated using the median and geometric standard deviations from the two parameter lognormal distributions that have been constructed for each site from a censored sample equivalent to that used for the hybrid model (ie  $\chi_{10}$ - $\chi_{50}$ ). Alternatively the observed mean can be related to the median through equation (5.7), using the maximum likelihood estimates of  $\beta_\chi$  for each site, and equation (5.5).

**Table 5-5:** Comparison of observed and HEC predicted annual average and maximum ground level concentrations of TSP,  $\mu\text{gm}^{-3}$ , adjusted for a background level of  $42\mu\text{gm}^{-3}$  crustal TSP

Site	Observed Mean	Predicted Mean	Expected Maximum	Predicted Maximum
Hamilton	55	57	191	205
Normanby	97	68	286	248
Petrie Bight	53	76	132	277
QIT	109	73	425	263
South Brisbane	58	67	123	249
W'gabba	99	73	265	263

The entire frequency distribution can also be generated from the geometric mean and standard deviation under the assumption of lognormality as shown in Figures 5-1 and 5-2. Any percentile relating to an appropriate air quality goal can then be selected. Figures 5-1 and 5-2 also demonstrate the good correlation between observed and predicted TSP concentrations, corrected for crustal background levels, at both sites.

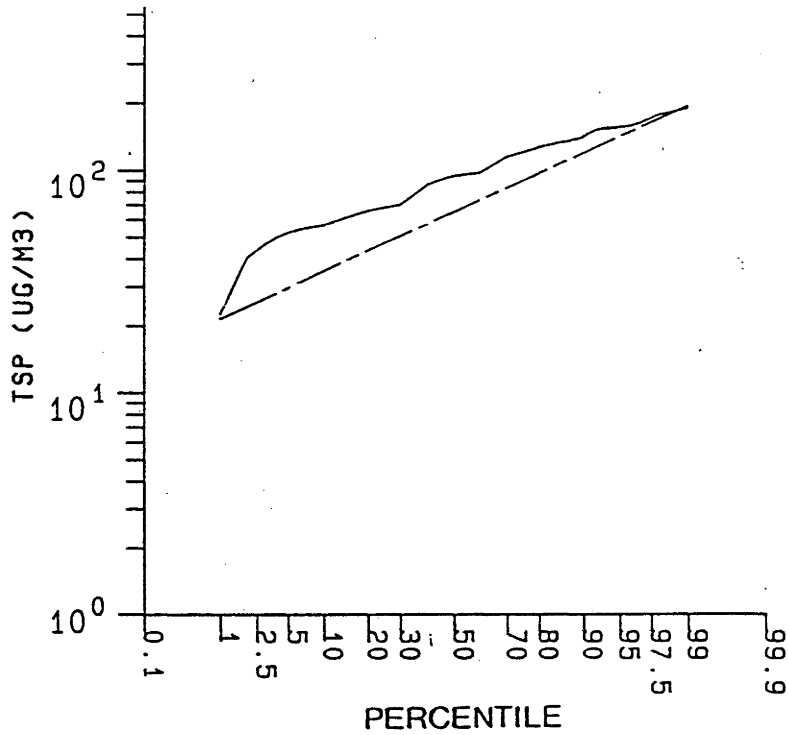
**5.4. CONTROLLING FOR THE MAXIMUM**

Chapter 4 identified that the mean guideline of  $90\mu\text{gm}^{-3}$  would be exceeded in 5 grid squares. These hot spots have been characterised in terms of the dominant sources and source types enabling appropriate control strategies to be developed. The maximum guideline of  $260\mu\text{gm}^{-3}$  chosen for use in this study, however, would be exceeded in 9 grid squares by the levels shown in Table 5-6. This is displayed graphically in Figure 5-3. Observed TSP levels are unavailable for the majority of grid squares and therefore it is difficult to fully validate these results. This can only be done through the expansion of the existing monitoring network. From the results in Table 5-5, however, it appears to be within acceptable limits of accuracy.

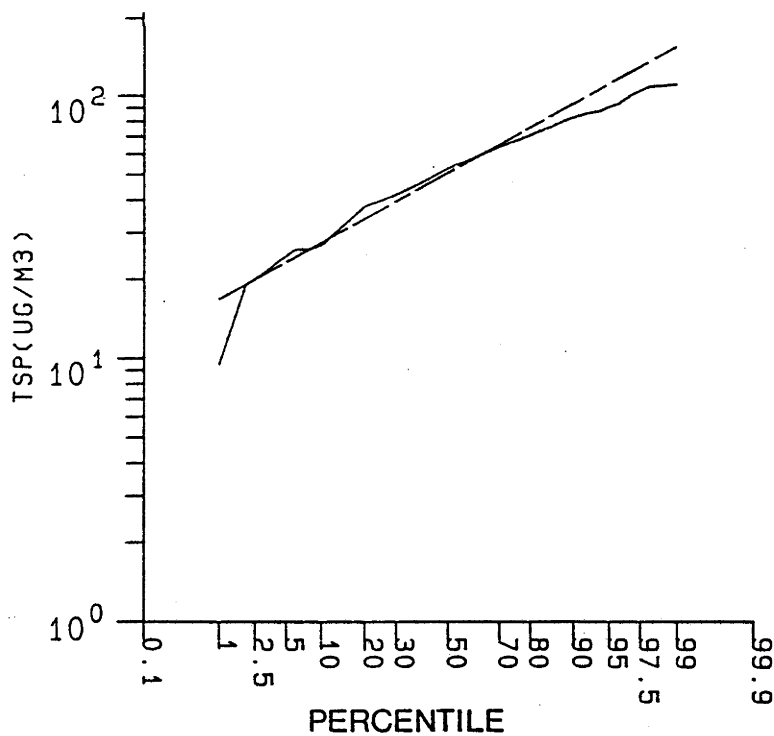
It can be seen from Table 5-6 that goals for short and long term exposure must be examined together if particulate emissions are to be appropriately controlled in the Brisbane region. Using the standard geometric deviation from the hybrid model to predict the upper percentile values of the frequency distribution it is suggested that the maximum goal of  $260\mu\text{gm}^{-3}$  would be exceeded in 4 grid squares where the annual mean TSP levels would remain within acceptable limits.

To help determine appropriate source controls, within a particular grid square,

**Figure 5-1:** HEC predicted concentrations corrected for background crustal concentrations against observed TSP concentrations at the Wolloongabba site, 1978-79

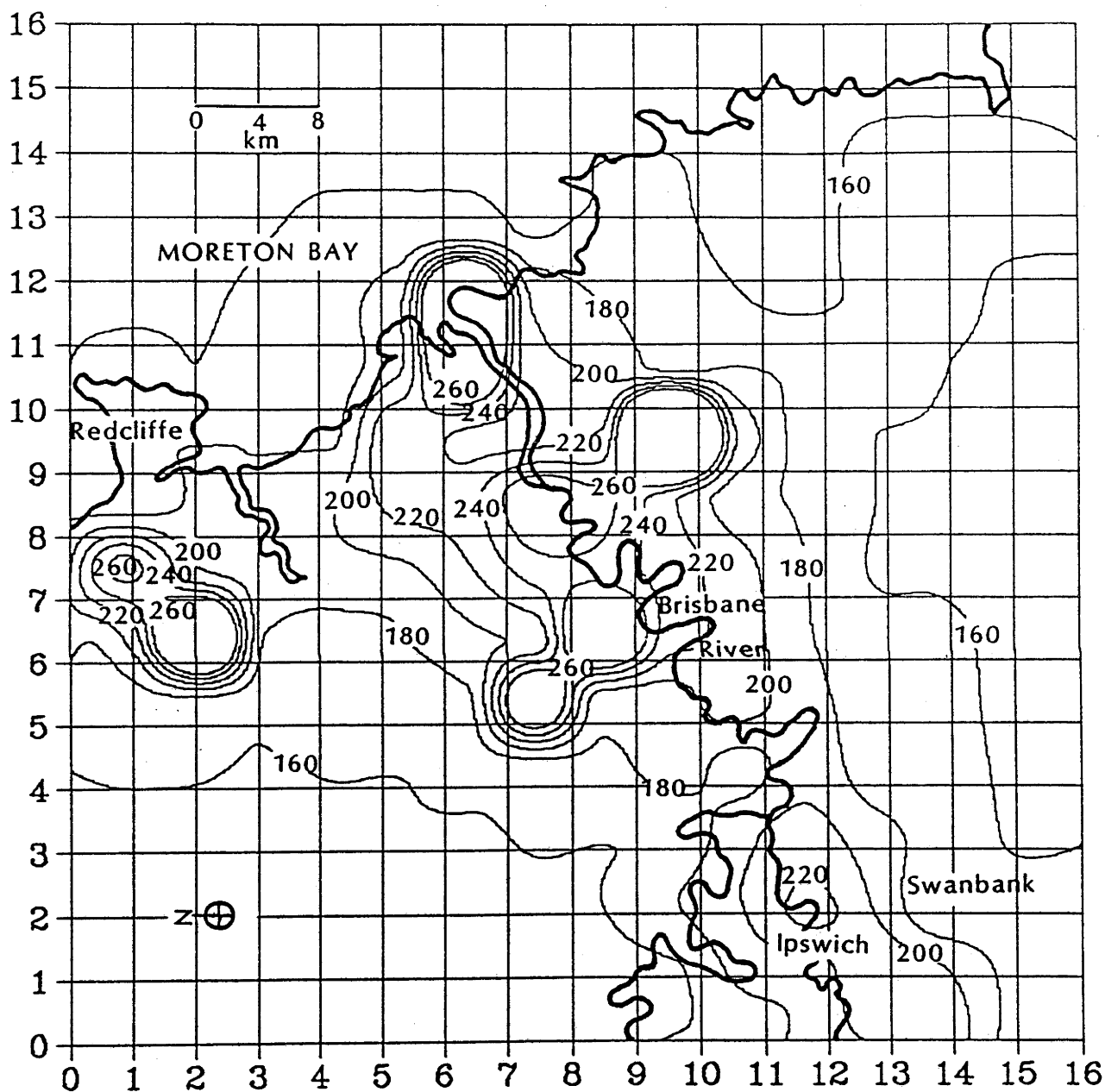


**Figure 5-2:** HEC predicted concentrations corrected for background crustal concentrations against observed TSP concentrations at the Hamilton site, 1978-79





**Figure 5-3:** Contour plot of annual average 24 hour maximum TSP,  $\mu\text{gm}^{-3}$ , from the HEC model output, adjusted for background crustal TSP, using UNIRAS Geopak routine GCONR2



**Table 5-6:** Ratios\* of area to point source contributions to ATDL predicted maximum ground level concentrations for grid squares where the USEPA 24 hour maximum standard for TSP is exceeded

Site	Predicted Mean $\mu\text{gm}^{-3}$	Predicted Maximum $\mu\text{gm}^{-3}$	Area to Point Source Ratio
Petrie Bight	77	280	0.87
Woolloongabba QIT	74	269	0.89
Lytton	108	392	0.94
Pinkenba	93	338	0.89
Holland Park	133	485	0.40
Petrie	103	372	0.47
Narangba	74	267	0.67
M <sup>t</sup> Cootha	91	330	0.57
Ferny Grove	85	308	0.52

\*Based on ATDL model output adjusted for crustal background

the ratio of area to point source contributions to the predicted ground level concentrations can be examined as for Chapter 4. Table 5-6 demonstrates that the ground level concentrations of TSP for the inner city sites of Woolloongabba/QIT and Petrie Bight are dominated by area sources. The ground level concentrations at the industrial sites of Lytton and Pinkenba are also largely attributable to area source emissions. Point source controls, however, would be more appropriate for the sites of Holland Park, Petrie, Narangba, M<sup>t</sup> Cootha and Ferny Grove. The Holland Park, Petrie and M<sup>t</sup> Cootha sources contributing to these violations of the maximum goal are the same as those for the mean (see section 4.3). The Narangba and Ferny Grove sites are also dominated by quarries.

Reductions in particulate emissions of approximately 24% by the year 1990 (Stewart et al, 1983), through the introduction of unleaded petrol, will improve ambient TSP maxima at all sites, as was the case for mean TSP concentrations. It is expected that violations of the USEPA 24 hour maximum standard will be less frequent with the introduction of unleaded petrol. The predicted maximum concentration for Woolloongabba would be reduced from 269 to an acceptable  $255\mu\text{gm}^{-3}$ . However, the Petrie Bight site would only be reduced from 280 to  $265\mu\text{gm}^{-3}$  which is still in excess of the USEPA 24 hour maximum standard.

### 5.5. APPLICABILITY TO CURRENT STRATEGIES

The ATDL model based on 1978/79 emissions was found to be applicable for the estimation of mean TSP concentrations up to 1984. A similar test has been conducted for the HEC model estimation of maximum TSP concentrations.

**Table 5-7:** Expected,  $\chi_E$ , and HEC predicted,  $\chi_p$ , maximum ground level concentrations of TSP,  $\mu\text{gm}^{-3}$ , 1979-84

Year	Variable	Hamilton	W'gabba	Darra	Jamboree	Pinkenba	Rocklea
1979	$\chi_E$	147	241	912	156	-	207
	$\chi_p$	199	256	206	203	-	203
1980	$\chi_E$	230	240	511	130	431	222
	$\chi_p$	177	222	189	177	177	177
1981	$\chi_E$	158	220	530	99	375	207
	$\chi_p$	305	408	305	310	290	300
1982	$\chi_E$	-	164	153	109	272	283
	$\chi_p$	-	290	221	221	214	214
1983	$\chi_E$	159	274	321	88	268	213
	$\chi_p$	206	272	213	213	199	206
1984	$\chi_E$	164	285	382	134	547	271
	$\chi_p$	266	357	280	267	267	271

The HEC model has been executed over the years 1979 to 1984. The 1978-79 emissions inventory and wind speed and direction data for each of these years have been used. When compared with the expected TSP levels 1979-84 in Table 5-7 the model provided predictions of maximum TSP concentration within a factor of 2 at most sites for most years. The notable exceptions to this are the years of 1981 and, to a lesser extent, 1984. This may be attributed in part to the values for  $\beta_u$  for these years being relatively high, 1.72 and 1.67 respectively. It is also noted that the emissions data for Darra are clearly incomplete with fugitive emissions from a large local source contributing to monitored TSP concentrations until 1982.

It has been shown in Chapter 4 that for the site of Woolloongabba the assumption that emissions have remained relatively constant is quite acceptable. It is not clear, however, whether the background crustal TSP concentration of  $42\mu\text{gm}^{-3}$  can be assumed to be constant for all years and sites. This reinforces the need to check calibration factors such as this whenever possible. Nonetheless, if

these assumptions hold then it is reasonable to suggest that the prediction of maximum TSP concentrations by this model is valid for the present day development and implementation of particulate emission control strategies in Brisbane.

## 5.6. DISCUSSION

The protection of human health from the effects of air pollution requires the control of short duration high concentration exposure as well as long term exposures of lower concentration. For this study the 24 hour maximum goal of  $260\mu\text{gm}^{-3}$ , established by the USEPA, has been selected as the concentration not to be exceeded for acceptable short term exposure. This has been combined with the Queensland DNAAPC long term annual arithmetic mean for TSP, used in Chapter 4, to provide a broader guideline to ambient air quality. The development and implementation of a control strategy based upon these goals has been examined using the TSP HEC model. The development of this model has extended the ATDL model and thereby provided an essential link between estimates of the upper percentile values of the frequency distribution and the actual emissions of a given pollutant.

In the process of developing the TSP HEC model development a simple hybrid model has been found to predict annual 24-hour maximum TSP concentrations to an acceptable level of accuracy based on total emissions. The hybrid model alone does not supply sufficiently detailed information for effective emission control decision-making. However, having satisfied the assumptions of the hybrid model developed for Brisbane the use of the geometric standard deviation for the inverse of wind speed to extend the ATDL model can be justified.

Extension of the ATDL model to include the output of other statistics from the frequency distribution for given TSP data has been shown to be relatively simple and provides acceptable results. This HEC model can predict the frequency distribution of ground level concentrations for TSP using emissions and meteorological data thereby providing estimates of both mean and maximum ambient concentrations. As these concentrations are linked to specific emissions they may be reviewed as part of existing control strategies. It has been found here that attempts to control the annual mean ground level concentration will not necessarily be sufficient to control the annual maximum.

As outlined in Chapter 4, point source emissions that contribute to violations of ambient air quality goals can be controlled through the licensing and review

procedures of the DNAAPC. The terms of reference for the DNAAPC, defined by the Clean Air Act (1962-1986), are limited to the prevention and minimisation of air pollution from industrial and commercial premises. Area source emissions generated by trucks, conventional motor vehicles and domestic activities are, therefore, beyond the legislated jurisdiction of the DNAAPC. The effect of unleaded petrol on area source contributions to ground level maximum concentrations are similar to that demonstrated for mean ground level concentrations in Chapter 4.

Information on the contribution of crustal material to ambient TSP has been necessary to produce this model. Although the estimates of background crustal TSP are crude, the performance of the model suggests that they are sufficiently accurate. It has also been shown that emissions have not changed substantially over the 1979-84 period and therefore conclusions relating to emissions control are of some relevance to current air quality management in Brisbane. It is important to acknowledge, however, that, like all atmospheric models, the predictions are constrained by uncertainties due to the stochastic nature of the processes being modelled. Errors arising from these fluctuations are of the order of a factor of two and this must be acknowledged when interpreting these predictions (Hanna, 1982).

The limitations in the Brisbane emissions inventory (Borowski, 1984) constrain the ability to validate the TSP HEC model. The use of crude background estimates, however, has enabled the identification of:

- areas of concern where emissions should be reduced to maintain air quality within acceptable limits
- maximum concentrations in excess of prescribed goals where the mean levels may be acceptable, and
- direct links between known sources and the violation of short and long term standards.

The prerequisite data for following the HEC approach are (1) meteorological data that may be combined with (2) ambient pollutant concentration data for the hybrid techniques and (3) emissions inventory data that can be used with the wind speed and direction data from (1) according to the ATDL model. Many regulatory authorities have emissions inventories for major population centres. Although not all contain particulate emission data, other inert pollutants of concern may be investigated using this technique. The potential exists, given an accurate emissions inventory, for the HEC approach to be successfully applied and the resulting models operated by regulatory authorities. This would enable decision-makers to control existing emissions and the introduction of new sources to urban areas in terms of both short and long term standards.

It has been shown how the ATDL and hybrid models may be linked simply and effectively if the appropriate assumptions are satisfied. A potential source of error in linking the hybrid and ATDL models is derived from assuming a lognormal distribution for the data where another may be more appropriate. This is investigated in the following Chapter along with an examination of the applicability of the TSP HEC model to other urban areas.

## Chapter 6

# UNCERTAINTIES AND GENERAL APPLICABILITY

### 6.1. INTRODUCTION

It has been shown that, when combined with estimates of background crustal concentrations, the Atmospheric Turbulence Diffusion Laboratory (ATDL) model can be extended to provide estimates of the annual maximum TSP concentration and other statistics from the frequency distribution for this pollutant in Brisbane, Australia. The usefulness of this modelling extension for the control of particulate emissions and urban air pollution has been discussed earlier and the questions must now be posed, what uncertainties can be quantified for the TSP HEC model and how applicable is this model to the problem of particulate air pollution in other urban areas in Australia?

The ATDL model is extended through the use of the hybrid modelling approach. As outlined in Chapter 4 this approach combines statistical and deterministic submodels to produce a hybrid model which estimates the entire distribution of pollutant concentrations within confidence limits. The first step of this approach requires a statistical distribution to be identified to represent the distribution of concentrations for the pollutant of concern. TSP data for the Brisbane urban area have been analysed and the two parameter lognormal distribution has been identified as the most appropriate statistical distribution to represent these data. Secondly, an appropriate shape parameter for this distribution must be identified. This parameter has been derived from an empirical relationship between TSP and wind speed data. The relationship has been empirically established for Brisbane and produces a shape parameter equivalent to the natural logarithm of the geometric standard deviation for the inverse of wind speed. When combined with the annual mean produced by the ATDL model it has been found that acceptable predictions of the entire frequency distribution for TSP data can be made in Brisbane. These predictions are then linked directly through models, such as the ATDL model, to the emissions input data and control strategies for point and area source emissions.

To examine the uncertainties in the hybrid component of this method and the potential for applying the TSP hybrid emissions control model to other urban areas, annual TSP and wind speed data sets have been obtained for major urban areas in Australia. In this Chapter the assumption of a two parameter lognormal distribution to represent the frequency distribution of annual TSP data sets in Australia is first tested. If this assumption is valid the question is then posed as to whether or not the shape parameter for this distribution, calculated from the inverse of wind speed, can be used to relate the annual mean for TSP to other statistics of the frequency distribution in urban areas other than Brisbane.

## 6.2. UNCERTAINTIES IN THE BRISBANE MODEL

Six years of TSP at 6 monitoring sites in Brisbane have been used for this evaluation. The "preferred distribution" in Table 6-1 is that selected by the maximum of the log-likelihood function. At these sites in Table 6-1 the differences between the estimated maxima using a preferred distribution (gamma or Weibull selected using maximum likelihood techniques) and those maxima that would be estimated by an assumed lognormal distribution have been illustrated. It can be seen from this Table that overprediction is likely in Brisbane, particularly if the lognormal distribution is assumed where the Weibull distribution is more appropriate. From the results in Chapter 5 extensive overprediction is not apparent. This supports the assumption of lognormality for the TSP data used in this study.

## 6.3. GENERAL APPLICABILITY

To examine the applicability of the approach the two primary assumptions are tested.

### The Lognormal Assumption

The first uncertainty concerning the general applicability of the TSP HEC model is the assumption of two parameter lognormality for all TSP data in Brisbane. It has been shown that if the lognormal distribution is assumed, where either gamma or Weibull distributions provide better estimates, then overprediction will occur in the Brisbane data.

Table 6-2 contains the percentage error, for other sites around Australia, that may be expected if a lognormal distribution is assumed to estimate the annual 24 hour maximum where another distribution, such as the two parameter Weibull or gamma, is more appropriate. A more detailed analysis of the distributional form of TSP and the errors in estimating the percentiles in Australian airsheds is currently



**Table 6-1:** Estimates of the maxima,  $\mu\text{gm}^{-3}$ , -95% confidence interval ( $\chi_p$ ) +95% confidence interval, derived assuming a lognormal distribution and compared with the preferred gamma or Weibull distributions

Preferred Distribution	Site	Year	Assumed Lognormal $\chi_p$	Preferred Distribution $\chi_p$
gamma	Hamilton	1978/79	146(197)270	102(148)192
	Hamilton	1979	133(176)235	101(142)181
	Woolloongabba	1983	208(273)365	153(224)294
	Rocklea	1983	146(192)257	108(156)206
	Darra	1984	265(405)578	198(290)380
	Jamboree Heights	1984	101(141)201	69(101)132
	Hamilton	1984	151(210)299	102(150)196
Weibull	Darra	1979	326(429)575	230(253)285
	Rocklea	1980	132(169)221	96(104)116
	Woolloongabba	1984	301(420)599	202(226)263
	Rocklea	1984	219(305)435	147(164)191

under investigation (DeWytt et al, 1987). For the sites and years tested it appears that the assumption of lognormality for TSP data is often acceptable. This assumption is clearly valid in Brisbane (91%), Sydney (100%) and Melbourne (100%) but less so in Adelaide (56%) and Canberra (64%). Previous research has found that the minimum error expected in estimating the 99<sup>th</sup> percentile using maximum likelihood techniques for a two parameter lognormal distribution (with sample size  $n=50$  and shape parameter  $b=0.5$  expressed as relative root mean square error) is 13.6% (Jakeman et al, 1986a). This provides a reasonable guide for the review of the absolute errors presented in Table 6-2 as maximum likelihood techniques have been used to estimate the annual maximum for the lognormal distribution of TSP concentrations. The sample sizes for these data sets varied from 40-60 and the average shape parameter for lognormally distributed TSP data was 0.47.

In Brisbane the absolute errors remain within a factor of two, an acceptable level of accuracy for air pollution models (Hanna, 1982). It is also notable that, by falsely assuming a lognormal distribution where in fact Weibull or gamma are preferred, resultant over-estimation of the maximum will subsequently lead to a conservative emissions control strategy. If this level of conservatism is acceptable then it may possible to apply the TSP hybrid emissions control model in Sydney and Melbourne using the assumption of lognormality for the particulate air

**Table 6-2:** Comparison\* of preferred distributions and errors in estimating the annual maximum from a two parameter lognormal distribution when another distribution is preferred gamma or Weibull

Australian Urban Areas	Brisbane	Sydney	Melbourne	Adelaide	Canberra
Number of Sites	6	5	8	3	4
Number of Years	1979-84	1981-84	1979-83	1979-82	1981-84
Preferred gamma % (no. of data sets)	16(6)	30(6)	33(6)	22(2)	14(2)
Preferred Weibull % (no. of data sets)	12(4)	10(2)	6(1)	44(4)	43(6)
Preferred lognormal % (no. of data sets)	71(24)	60(12)	61(11)	33(3)	43(6)
Accepted lognormal <sup>1</sup> %	91	100	100	56	64
Error estimating lognormal maximum if preferred gamma %	20-40	13-25	12-30	24-161	32-37
Error estimating lognormal maximum if preferred Weibull %	70-85	36-47	41	57-122	70-215

\* only annual TSP data sets containing >66% of a maximum yearly sample (N=60) have been used for this analysis

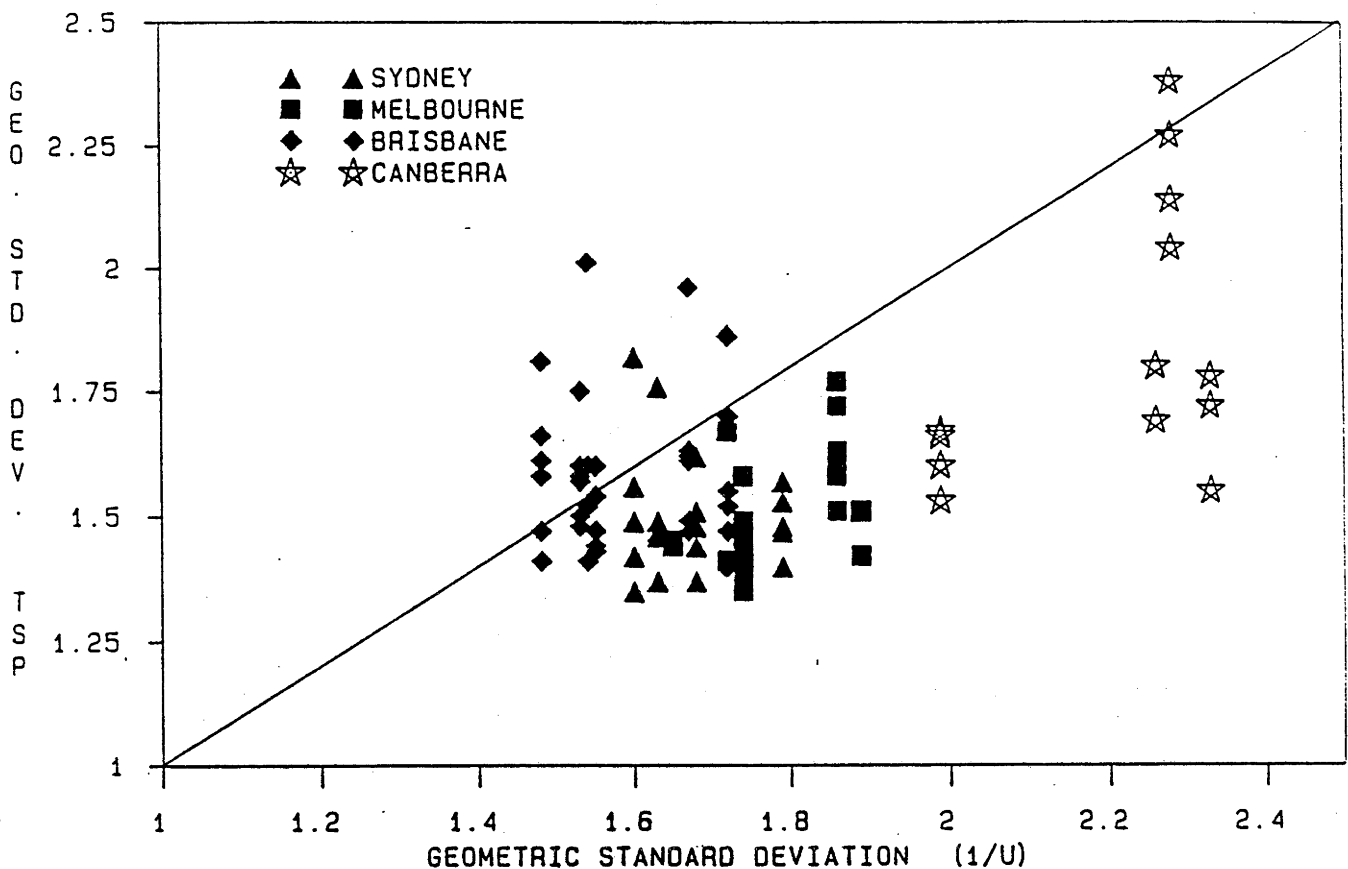
<sup>1</sup>some data sets are accepted as lognormal using a Kolmogorov Smirnov distributional test (see section 5.2.1) although they are preferred gamma or Weibull by testing for the maximum of the log-likelihood functions (Taylor et al, 1986) pollution data. Sites in Adelaide, however, appear to produce much higher errors when estimating the annual maximum if the assumption of lognormality is invalid. Canberra produces the highest errors associated with an invalid assumption of lognormality. It should also be noted that a smaller percentage of sites in these latter two urban areas are preferred lognormal.

### The Empirical Assumption

The assumption that air pollutant percentiles may be described by the relatively simple inverse relationship defined in equation (3.5) has been examined on a number of occasions (Simpson et al, 1986, Simpson et al, 1987, Taylor, 1985). If valid this relationship can then be used to estimate the shape parameter for the lognormal distribution. From equation (3.5) the shape parameter is given directly by the inverse of wind speed. For a lognormal distribution this shape parameter is the logarithm of the geometric standard deviation ( $\beta$ ).

Figure 6-1 compares the geometric standard deviations, generated from two parameter lognormal distributions, for annual TSP data,  $\beta_{TSP}$ , with the corresponding annual inverse wind speed data,  $\beta_u$ . The diagonal axis represents the 1:1 relationship between  $\beta_{TSP}$  and  $\beta_u$ . It can be seen from this figure that there is

**Figure 6-1:** Geometric standard deviations for the lognormal distributions estimated from TSP and inverse wind speed data at all sites over all years (excluding Adelaide)



no obvious relationship between  $\beta_{TSP}$  and  $\beta_u$  that is consistent for all sites. The points are well spread but it is noteworthy that the scatter for the sites in the Brisbane area produced acceptable predictions using the TSP HEC approach.

There is an apparent clustering of the east and south eastern coastal urban areas of Brisbane, Sydney and Melbourne (wind speed data have not been collected for Adelaide). This grouping does not overlap with Canberra data. This is supported by an examination of average geometric standard deviations for each urban area shown in Table 6-3. It suggests that Canberra as an inland city, may be distinct in terms of annual wind speed variability from the coastal areas of Brisbane, Sydney and Melbourne and should, therefore, be treated differently. Other factors are also likely to enhance this distinction (eg source types and topography). It appears that overprediction will occur if  $\beta_u$  is used to extend a model like the ATDL model for most sites in Sydney, Melbourne and Canberra. This will produce a conservative emissions control strategy for particulates.

The question may be asked whether an average  $\beta_{TSP}$  value may be used for the eastern and south eastern coastal sites. From Figure 6-1 it appears that an upper

**Table 6-3:** Comparison of average geometric standard deviations for the urban areas of Brisbane, Sydney, Melbourne and Canberra

Parameter	Brisbane	Sydney	Melbourne	Canberra
TSP $\beta_{\pm sd}$ (N)	$1.580 \pm 0.152$ (34)	$1.503 \pm 0.120$ (20)	$1.514 \pm 0.117$ (20)	$1.825 \pm 0.272$ (14)
Inverse Wind speed $\beta_{\pm sd}$ (N)	$1.582 \pm 0.092$ (6)	$1.675 \pm 0.084$ (4)	$1.772 \pm 0.100$ (5)	$2.220 \pm 0.153$ (4)

and lower bound may exist for the variability of  $\beta_{TSP}$ . Since Canberra should be treated separately from Brisbane, Sydney and Melbourne, an average  $\beta_{TSP}$  has been calculated only for these latter sites. The predictions of the annual maxima for TSP using this geometric standard deviation ( $\beta_{TSP}=1.541$ ) are compared with the  $\beta_u$  predictions for lognormally distributed TSP in Table 6-4.

**Table 6-4:** Comparison of % absolute errors derived when estimating the lognormal annual maxima using an average TSP geometric standard deviation,  $\beta_{TSP}$ , and the geometric standard deviation,  $\beta_u$ , calculated from the inverse of wind speed

Geometric Standard Deviation	Brisbane	Sydney	Melbourne
$\beta_{TSP}$ average % error  $\pm sd$ (% range)	$18.9 \pm 14.3$ (1.0-54.7)	$20.3 \pm 13.7$ (2.3-49.2)	$21.78 \pm 11.6$ (6.1-49.4)
$\beta_u$ average % error  $\pm sd$ (% range)	$26.3 \pm 19.8$ (1.9-61.0)	$48.2 \pm 26.7$ (5.8-108.4)	$69.6 \pm 36.0$ (9.6-137.8)

Table 6-4 demonstrates that on average the percentage absolute errors associated with using a geometric standard deviation, derived from the inverse of wind speed, tend to be higher than if a constant geometric standard deviation, derived from an average of TSP geometric standard deviation, was used. These differences, however, are not statistically significant due to the high variance to mean ratio.

## 6.4. DISCUSSION

In Brisbane the TSP hybrid emissions control extension of the ATDL and hybrid models, has been used to predict statistics from TSP frequency distributions in addition to the annual arithmetic mean and has yielded acceptable results (see Chapters 4 and 5). Uncertainties exist about the general applicability of the hybrid modelling technique used to extend a model, such as the ATDL model, in Brisbane and other urban areas. The assumptions of the TSP hybrid emissions control model, that (1) the frequency of total suspended particulate data can be reliably represented by a two parameter lognormal distribution; and, (2) a simple deterministic relationship between percentiles of pollutant concentration and the inverse of wind speed exists for this urban area, were tested at a number of locations in Australia and over a number of years.

One potential source of error in applying the TSP hybrid emissions control model is derived from assuming a lognormal distribution for the data where another may be more appropriate. The lognormal assumption (1) is shown to be valid for most sites and years. The errors that may be expected if the two parameter lognormal distribution is inappropriate have been estimated. The errors at particular sites around Australia, if the assumption of lognormality is invalid, vary in magnitude. Inappropriately using a two parameter lognormal probability density function to estimate the maximum TSP concentration produces errors in Sydney, Melbourne and Adelaide comparable to those for Brisbane. If invalid the lognormal model may over-estimate the maximum concentration by between 12 and 161% where the two parameter gamma distribution is preferred and between 36 and 122% where the two parameter Weibull distribution best represents the statistical distribution of TSP concentrations at these sites.

In Canberra, however, the estimated maximum may be up to a factor of three greater than the observed. This reinforces the need to carefully execute step (1) of the hybrid modelling approach in order to clearly identify the statistical distributions for the pollutant concentrations. It is interesting to note that the lognormal assumption is less acceptable at the sites of Adelaide and Canberra. These sites may be geographically and meteorologically distinct with potentially different emissions regimes. More detailed work is required before any reasonable conclusions of this nature can be confirmed. If these apparently distinct sites are reviewed separately then the largest percentage errors found under an invalid lognormal assumption when estimating the annual maximum in Brisbane, Sydney and Melbourne are 85% and 40% for Weibull and gamma distributions respectively. The lognormal model consistently over-estimates the maximum compared with the Weibull and gamma distributions and, therefore, overall these results indicate that the lognormal distribution may be a good conservative assumption for air quality management.

The empirical assumption that urban TSP concentrations are inversely proportional to wind speed has been tested for the same sites (except Adelaide) and years. A comparison of geometric standard deviations for TSP and the inverse of wind speed demonstrates that this model may be as successful at some urban areas on the eastern coasts of Australia as it has been in Brisbane. Again it is suggested that the application of this model would tend to over-estimate the upper percentile statistics of the frequency distribution using the geometric standard deviation for inverse wind speed as the exponentiated shape parameter for that distribution.

These assumptions have not been tested at industrial locations where point source emissions will dominate the ground level concentrations that are monitored. Under these conditions other variables, such as wind direction, become important. It has been suggested that sites dominated by point source emissions are often best represented by the Weibull distribution (Jakeman et al, 1987a). This study indicates that the largest errors through overprediction would be found if the lognormal distribution was assumed under these conditions.

Assumption (2) of the hybrid approach appears to be the weakest. It is questionable whether wind speed statistics alone will satisfactorily explain the variance in the air pollution data as suggested by this assumption. Indeed, the performance of the TSP HEC model would seemingly be improved if an average TSP geometric standard deviation is used to replace geometric standard deviation based on wind speed statistics. This result is not unexpected as variability in emissions strengths throughout the year and the siting of wind speed monitors are likely to be important. For the TSP HEC model, emissions rates have only been available as annual averages used to predict the annual mean (eg using the ATDL model). One cause of variability in emissions would be seasonal cycles. Canberra is a city in which particulate emissions have a seasonal cycle due to a large wood fire component (Taylor et al, 1987). The other urban areas examined have a distinct industrial emissions component that is absent in Canberra. Industrial emissions tend not to exhibit seasonal variation and wood fires in these other cities are far less important (Newman, 1982).

Although the results presented here provide only a rough guide as to the potential applicability of the TSP HEC model, it is suggested that, for a city in which particulate emissions are dominated by sources with little marked seasonal variability (eg cars and industry), the TSP hybrid emissions control model appears to be generally applicable for estimating TSP distributions from emissions and meteorological data. Both assumptions appear to be useful in this application as they would tend to produce a conservative control strategy for particulate emissions. The HEC model performance may be improved if the mean value for the geometric standard deviations of TSP concentrations, based on averaging  $\beta_{\text{TSP}}$  for all east and south east coast sites for all years, is used. It is clear, however, that more work in this area is warranted.

The next question to be addressed is whether or not TSP is an adequate measure for relating particulate concentration to human health? The evidence of health effects suggests that measurement of the ambient fine particulate matter less than  $10\mu\text{m}$  ( $\text{PM}_{10}$ ) would be more appropriate (Vate et al, 1986). Further investigation

of the relationship between TSP and  $PM_{10}$  in Brisbane is necessary to determine the applicability of the HEC model to health standards based on fine particulate concentrations.

## Chapter 7

# TOWARDS AN EMISSIONS CONTROL STRATEGY FOR INHALABLE AND RESPIRABLE PARTICULATES

### 7.1. INTRODUCTION

The TSP HEC model has been developed to relate violations of ambient air quality standards directly to emissions. The air quality goals in Brisbane are in terms of TSP, therefore, this measure has been used to validate the model. While the model based on this measure offers advantages for the management of airborne particulate matter by the DNAAPC, it must be assumed that this is an appropriate goal for the protection of human health. From reviewing the health concerns of particulate matter in Chapter 2 the evidence suggests that measurement of the ambient inhalable and respirable particulate matter less than  $10\mu\text{m}$  ( $\text{PM}_{10}$ ) may be more appropriate.

It has been shown in Chapter 4 that, when combined with estimates of background crustal concentrations, the Atmospheric Turbulence Diffusion Laboratory (ATDL) model provides accurate estimates of annual mean ground level TSP concentrations. In Chapter 5 the ATDL model is extended to produce the TSP HEC model which provides estimates of both annual averages and 24 hour maximum TSP concentrations. It has been shown that the TSP HEC model can be used in the development and implementation of control strategies to account for both short and long term goals for particulate pollution when measured as TSP. The question must however be asked, whether acute and chronic health effects of particulate air pollution are best related to TSP and subsequently how stringently the emission controls suggested by the TSP HEC model should be enforced.

In this Chapter, therefore, the health relevance of TSP and subsequently the TSP HEC model predictions are examined and the development of an emissions control strategy for inhalable and respirable particulates is investigated. By combining health concerns with current knowledge of particulates in Brisbane the TSP HEC model is related to health relevant measures for inhalable particles. The possibility



of using the HEC modelling approach to predict ground level concentrations of  $PM_{10}$  and subsequently control emissions of inhalable and respirable particulates will be investigated.

## 7.2. HEALTH RELEVANT MEASURES OF PARTICULATE AIR POLLUTION

This section examines whether TSP is an appropriate measure with respect to the health concerns of airborne particulate matter. Air quality goals and standards for TSP have been developed and used throughout the world. This largely because TSP has been found to provide a good aggregate measure of coarse, inhalable and respirable particulates commonly of the range  $0.3-100\mu m$ . It is now becoming clear that important information is lost in this aggregation and that essential health relevant data needs to be obtained for inhalable and respirable particles independently of total mass (Vate et al, 1986). This is reflected in the development of ambient air quality standards for  $PM_{10}$  by the USEPA and the increased level of  $PM_{10}$  monitoring in both the United States and Australia.

The health effects associated with TSP have been well documented in previous studies (Subcommittee on Airborne Particles, 1979) and more recently by Ware et al, 1986, Spinaci et al, 1985 and Kim, 1985. In these epidemiological studies TSP seems to be providing a good gross measure of ambient air quality rather than a direct relationship between specific inhaled particles and related health effects. It has been suggested by Lippmann and Liroy (1985) that the relationships between health effects and particulate pollution, based on epidemiological studies over the last 20 years, must be seriously questioned. Most of these studies are considered to be flawed through not accounting for non-ambient exposures, individual levels of activity and responsiveness, and short term versus long term exposure effects (Lippmann and Liroy, 1985).

As the primary physiological concerns for human health associated with particulate air pollution are derived mainly from the effects after ingestion, the size fraction of interest is predominantly particles less than  $10\mu m$  (see section 2). After ingestion the shape and chemical composition must be taken into account. To monitor particulate air pollution to account for these health considerations it is clear that both size and chemical composition must be included in the analysis.

Size selective inlets (SSI) for high-volume samplers to monitor  $PM_{10}$  are being used by the USEPA. This provides an aggregated measure of inhalable and respirable particulates and is designed to approximate inhalation through oral

breathing (Wedding and Weigand, 1985, Chan and Lippman, 1980). Standards have recently been developed by the USEPA for SSI  $PM_{10}$  monitoring with a long term annual arithmetic mean of  $50\mu g m^{-3}$  and a short term annual 24 hour maximum of  $150\mu g m^{-3}$  not to be exceeded. Chemical composition is known to vary considerably between different size fractions and consequently to estimate the potential health impact of soluble matter in the respirable range this size fraction must be collected and analysed.

In Australia dichotomous sampling has been used in Victoria by the EPA to provide information on the respirable fraction. Using 50% effective cut-points at  $10\mu m$  and  $2.5\mu m$ , this type of monitoring provides weight and concentration measurements for both the inhalable and respirable size fractions that can then be chemically analysed. Problems with maintaining the sample integrity of the respirable fraction are known to exist with this technique. Beta scattering monitored using nephelometry could give a continuous measure of this fraction as it has been found to correlate well with dichotomous samples  $<2.5\mu m$  (Bardsley et al, 1986). A one day in six sample using dichotomous samplers may then be sufficient to estimate the chemical composition.

### 7.3. INHALABLE AND RESPIRABLE PARTICULATE MATTER

There is a need to improve our understanding of  $PM_{10}$  in an urban environment for reasons of both health relevancy and the development of an appropriate control strategy. In this section the information base for inhalable and respirable particulates is examined in Brisbane. From this information some requirements for a health relevant control strategy for particulates can be identified in the Brisbane urban area.

Previous studies have found the problem of particulate air pollution in an urban environment to be very complex (Taylor et al, 1987, Carras et al, 1983). The inherent complexity and differences in each urban area can be attributed to variable emission types, locations and strengths; meteorology; size distributions and associated chemical and reflecting properties; and the chemical interactions between particles and other pollutants such as  $SO_2$ ,  $NO_x$  and  $O_3$ . These factors must be acknowledged when compiling the information available for inhalable and respirable particulates in the Brisbane airshed.

It is recognised that particulate air pollution is an air quality concern in Brisbane (Division of Noise Abatement and Air Pollution Control, 1986), as it is elsewhere in Australia (Department of Arts, Heritage and Environment, 1985), with a

considerable proportion (ie approximately 30%) of the resources of the DNAAPC utilised to monitor and control particulate air pollution in general (Nimmo, 1986). This is reinforced by the predictions of TSP concentrations made in Chapters 4 and 5. Recognition of the need to investigate the levels of inhalable particulates in Brisbane has led to SSI PM<sub>10</sub> being monitored routinely since 1986 at two sites, Wolloongabba and Rocklea, concurrently with high-volume TSP sampling. Light scattering has been monitored since 1979 using MRI 1591 and 1561 nephelometers at three sites (Eagle Farm, MRI 1591; Fortitude Valley, MRI 1591; and Rocklea, MRI 1561). This has provided information on visibility reduction due to airborne particulate matter that is dominated by particles <2 $\mu$ m. Dustfall has been monitored since approximately 1971 but represents only particles predominantly >20 $\mu$ m. As this size range is of limited health concern it will not be discussed here.

The information obtained from this network is supplemented by specific studies into the nature of particulate air pollution. During the winter months of 1985 a specific study of chemical composition was conducted for TSP (Verrall et al, 1986). It was found that there is a relatively high crustal component, between 40-64%, that contributes to the weight of TSP. This crustal profile was defined as containing elements of silica, aluminium, iron, chlorine, magnesium, sodium, calcium, copper, lead, potassium, phosphorus, titanium, zinc and sulphur. The relative proportions of these elements varied from site to site but silica and aluminium were found to consistently dominate the profile by weight. Other dominant source profiles included marine aerosols, a cement factory and motor vehicle emissions. The contribution of each of these to TSP levels varied between sites. A random selection of samples collected during this study was further analysed using light, scanning and transmission electron microscopy and electron micro-probe techniques at the Centre for Resource and Environmental Studies, Australian National University. This has enabled Brisbane TSP samples to be characterised in terms of size, shape and basic composition.

It was found from the results of the light microscopy shown in Table 7-1 that there was high variability from day to day between the proportion of particles within various size fractions. Due to this variation it is difficult to reach any statistically confident conclusions for the level of resolution provided by light microscopy. However, observations that can be made include:

- the mineral fraction is low, as a percent by number, compared with the high proportion by weight determined from chemical analyses;
- there is a high proportion of PM<sub>10</sub> averaging approximately 70% over all sites, although the variation in this proportion is high; and,

**Table 7-1:** Average %TSP by number  $\pm$ sd for dominant size fractions from microscopic analyses of Brisbane sites

Sites	Black Amorphous		Fine <3 $\mu$ m	Mineral		Total Inhalable <10 $\mu$ m
	3-10 $\mu$ m	10-30 $\mu$ m		3-10 $\mu$ m	10-30 $\mu$ m	
Pinkenba	37.9 $\pm$ 30.1	11.5 $\pm$ 7.4	22.7 $\pm$ 12.2	9.0 $\pm$ 11.5	2.3 $\pm$ 2.5	69.6
Rocklea	30.9 $\pm$ 15.9	26.8 $\pm$ 15.7	20.3 $\pm$ 17.6	3.3 $\pm$ 3.1	5.6 $\pm$ 4.0	54.5
Hamilton	22.2 $\pm$ 4.9	9.5 $\pm$ 10.8	43.2 $\pm$ 15.1	4.1 $\pm$ 6.0	1.3 $\pm$ 2.3	69.5
Jamboree Heights	35.0 $\pm$ 26.9	4.8 $\pm$ 7.2	9.7 $\pm$ 4.9	16.0 $\pm$ 19.0	1.3 $\pm$ 2.3	60.7
W'gabba	47.3 $\pm$ 21.8	16.2 $\pm$ 15.5	18.0 $\pm$ 16.4	12.0 $\pm$ 20.7	3.2 $\pm$ 3.5	77.3
Darra	24.3 $\pm$ 7.4	9.0 $\pm$ 6.2	49.7 $\pm$ 9.5	9.6 $\pm$ 16.7	2.0 $\pm$ 2.0	83.6

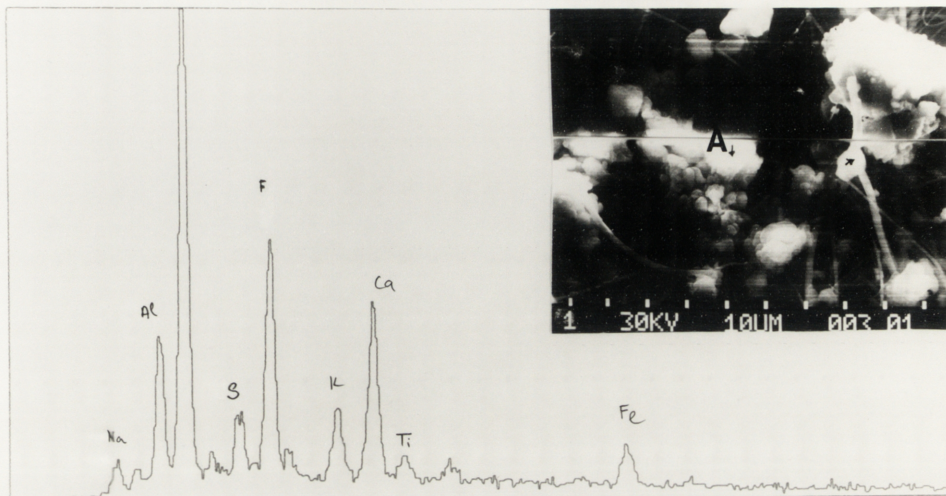
- between 16% and 60% of the PM<sub>10</sub> may be interpreted as respirable.

Transmission electron microscopy (TEM) and scanning electron microscopy (SEM) plus electron micro-probe, following methods detailed in Glikson et al (1987), have been used to provide further detail for the respirable fraction. Of the three days randomly selected from the chemical analysis study, days 196 and 239 were selected for the Darra and Woolloongabba sites respectively for SEM and electron microprobe analysis. Specific sections from these days, identifying specific properties of particles in the respirable range, are presented in Plates 7-1, 7-2 and 7-3.

Crustal material, characterised largely by silica (Si), aluminium (Al) and iron (Fe), can be found in the respirable range at both sites shown in Plate 7-1 and Plate 7-2. This supports similar findings in both Australia and the south west of the United States (Bardsley et al, 1986, Pitchford et al, 1981). Titanium (Ti), as a product of mineral sands operations detected by the chemical composition study (Verrall et al, 1986), is another crustal element that is found using SEM to be in the respirable range at both sites. Although less frequent than for a similar analysis of respirable particles in Canberra (Glikson et al, 1987), lead (Pb) and bromine (Br) associated with motor vehicle exhausts were also shown in Plate 7-3 to exist at Woolloongabba. Other elements that can be associated with specific sources include calcium (Ca), probably from cement works, and components of a marine aerosol including sodium (Na), chlorine (Cl) and potassium (K). All these and other elements have been isolated by Bardsley et al (1986) at sites in



**Plate 7-1:** SEM electron micro-probe of particle A;  
Darra, day 196, 1985



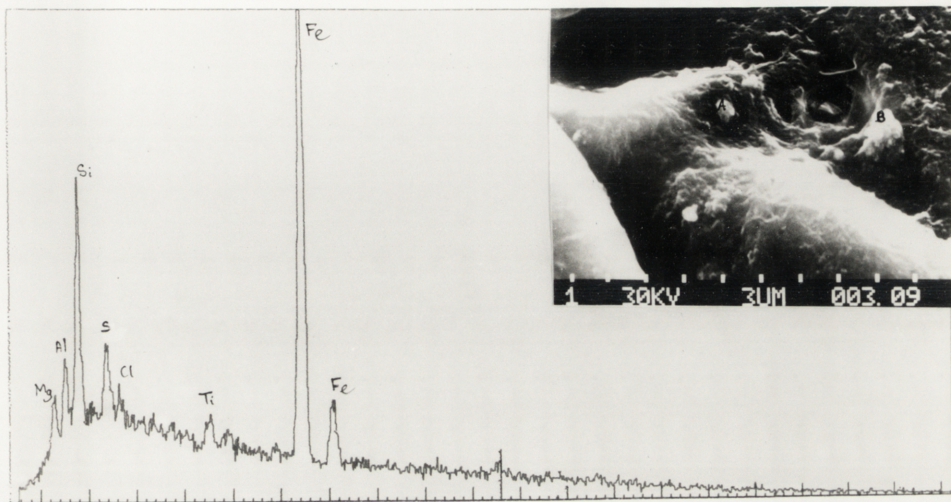
Melbourne using x-ray fluorescence analysis of dichotomous samples  $<2.5\mu\text{m}$ . The results of these two studies are not directly comparable as the micro-probe analysis is of individual particles as opposed to the total respirable mass examined in Melbourne.

The specific health concerns that are highlighted by the SEM micro-probe analysis relate primarily to Pb and sulphur (S). As outlined in section 2.2 these elements, individually and in association with other particulates, have been found to adversely effect human health at certain concentrations. The concentrations cannot be determined in this study but it is confirmed that they are found in the respirable fraction, see Plate 7-3. It should also be noted that, although crustal matter has been found to dominate TSP by mass, individual particles within the respirable range, shown in Plates 7-2 and 7-3, may be dominated by other elements such as Fe and S.

The site of Rocklea, during 1986, has been selected for a more detailed analysis



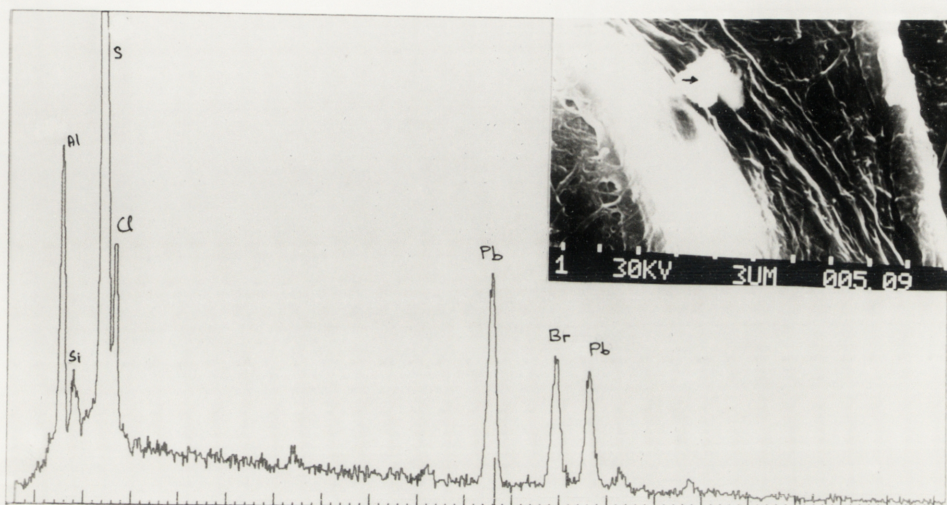
**Plate 7-2:** SEM electron micro-probe of particle A;  
Woolloongabba, day 239, 1985



of the relationships between TSP and SSI  $PM_{10}$ . This site has been chosen as it is the only location where these measures are sampled concurrently. Unfortunately the 1986 light scattering data is invalid due to instrument malfunction. In Figure 7-1 it is shown that, at this site, there is no apparent seasonal trend in the two measures of TSP and  $PM_{10}$ . There is, however, considerable variation throughout the year due largely to varying emissions and meteorological characteristics. What variations do exist seem to be reflected similarly in both TSP and SSI  $PM_{10}$  measurements. A least squares regression of these two parameters is found to be significant ( $p < .05$ ;  $T = 34.84$ ,  $t_{.025, 56}$ ) accounting for 77% of the variation (see Figure 7-2). From this regression analysis it was found that  $SSI\ PM_{10} = 0.50(TSP)$ . This supports earlier analyses conducted by the DNAAPC at a Brisbane urban site found the average ratio of TSP to  $PM_{10}$  to be approximately 0.49 (Woodland, 1985). The relationship established between TSP and  $PM_{10}$ , monitored using dichotomous samplers, at 8 sites from the Inhalable Particulate Network in the United States of America (Rodes and Evans, 1985) was also found to be similar. Rodes and Evans (1985) found that on average the ratio between  $PM_{10}$  and TSP



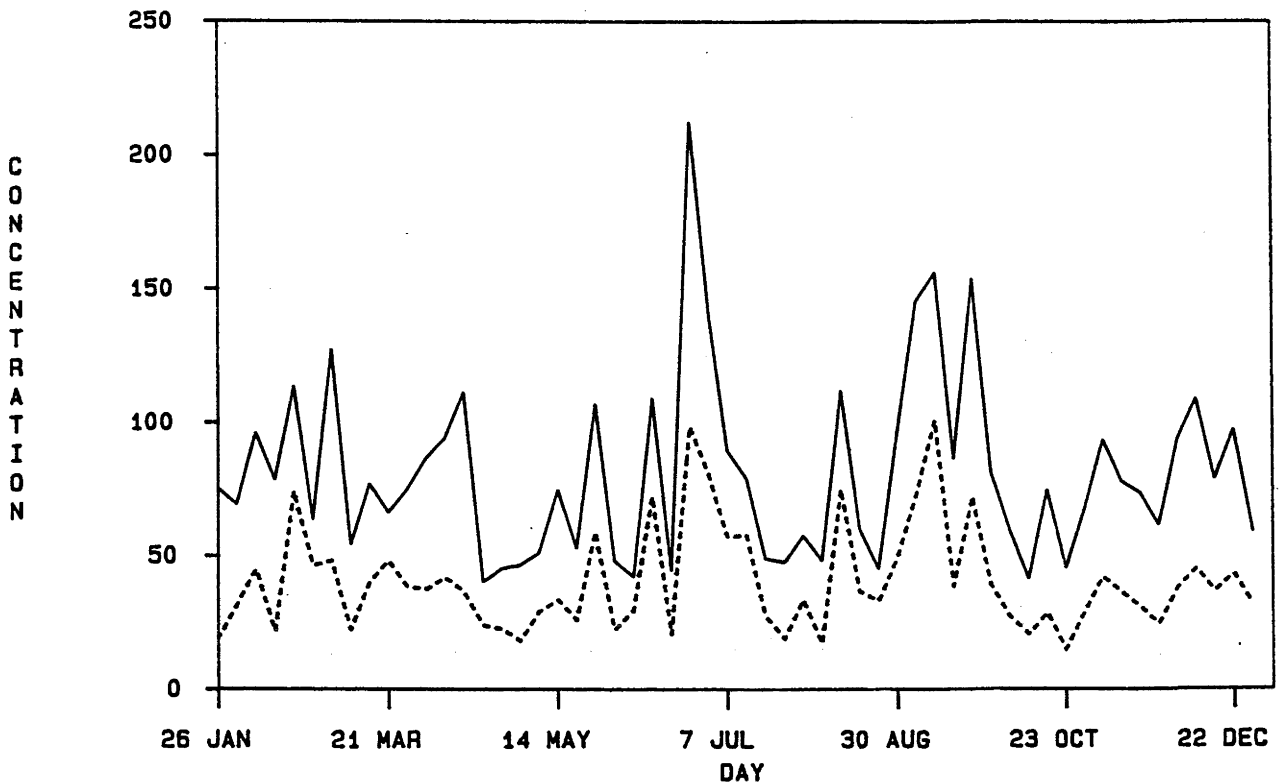
**Plate 7-3:** SEM micro-probe of a particle shown by the arrow;  
Woolloongabba, day 239, 1985



was 0.485. It should be noted that dichotomous, compared with SSI sampling, tends to underestimate the concentration of  $PM_{10}$  (Bardsley et al, 1986). From previous research it has been found that TSP does not correlate well with light scattering data measured as a beta scattering coefficient due to the properties and processes for respirable particles being quite distinct from coarse particles which dominate TSP (Subcommittee on Airborne Particles, 1979).

It is also interesting to note that the difference between TSP and SSI  $PM_{10}$  appears to be similar to the crustal component identified by chemical composition analyses. For the site of Rocklea, during the chemical composition study in 1985, it was found that the crustal proportion was approximately 0.59(TSP) by weight. As crustal matter often dominates the coarse particle fraction (Verrall et al, 1986, Vate et al, 1986, Subcommittee on Airborne Particles, 1979), the difference between TSP and  $PM_{10}$  may therefore be largely accounted for by crustal source emissions. This assumes that 1985 TSP levels are on average comparable with 1986 results. The results in Table 7-1 also indicate that the mineral fraction less than  $10\mu m$  is usually much less than other fractions.

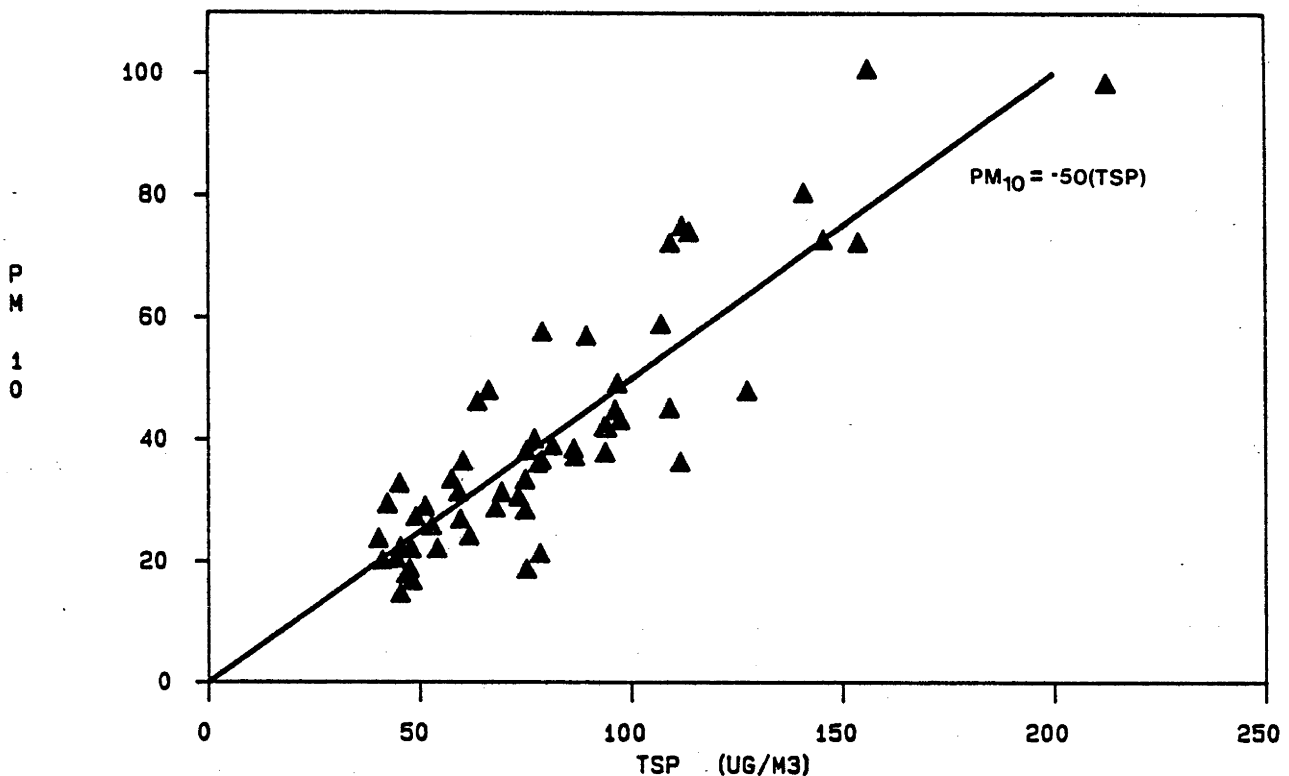
**Figure 7-1:** Annual variation in TSP and SS1 PM<sub>10</sub>  
at Rocklea, 1986



As detailed earlier the emissions inventory consists only of anthropogenic emissions and therefore does not account for a large proportion of crustal particulate matter. As no particle size differentiation is made in the inventory it can not be used directly to model inhalable and respirable particulates in the same way as has been done for TSP. It may be possible, however, to produce crude estimates for inhalable and respirable particulates using the HEC modelling approach if the relationship between TSP, PM<sub>10</sub> and crustal material is consistent throughout the urban area. It must also be assumed that all important sources of coarse particles can be removed from the emissions inventory. Before this can be attempted the assumptions of the HEC modelling approach must be examined for inhalable and respirable particulates (ie for PM<sub>10</sub> data).



Figure 7-2: Least squares regression for TSP *versus* SSI PM<sub>10</sub>,  
 $\mu\text{gm}^{-3}$ , at Rocklea, 1986



#### 7.4. THE HEC APPROACH FOR INHALABLE AND RESPIRABLE PARTICULATES

In this section the HEC modelling approach is examined to determine how it may be used to assist in the development and implementation of control strategies for inhalable and respirable particulates. The TSP HEC model has been developed from a general approach that is described in Figure 3-1. This figure illustrates how a deterministic model based on emissions and meteorological data, such as the ATDL model, can be used to predict the annual arithmetic mean and, when combined with the shape parameter for the lognormal distribution, it is then possible to estimate the entire frequency distribution of pollutant concentrations. From this distribution the number of violations of a given standard or goal (eg mean, median, maximum, 98<sup>th</sup> percentile) can be estimated. With this approach any model may be used to make the link between the estimated frequency distribution and emissions.

The TSP HEC model is based on the assumptions that (1) the frequency of total suspended particulate data can be reliably represented by a two parameter lognormal distribution; and, (2) a simple deterministic relationship between percentiles of pollutant concentration and the inverse of wind speed exists for this urban area. These assumptions have been found to be reasonably satisfied for TSP in other coastal urban areas of Australia such as Sydney and Melbourne. The hybrid modelling approach can also be used under different meteorological and emission regimes and yield different empirical and statistical assumptions (Jakeman et al, 1987a). The validity of these assumptions for inhalable and respirable particulates can be examined as a means of determining the applicability of the HEC method to these size fractions.

Table 7-2 shows the results of identifying statistical distributions for the various data sets. At Rocklea in 1986 it was found that the distributions of both TSP and SSI  $PM_{10}$  concentrations were best represented by a two parameter lognormal distribution. Both data sets therefore satisfy assumption (1). Given the good correlation between TSP and SSI  $PM_{10}$  this is not unexpected.

**Table 7-2:** Analysis of appropriate statistical distributions for TSP, SSI  $PM_{10}$  and light scattering at Rocklea

Ambient Particulate Measure	Preferred 2-parameter Distribution	Accepted by Kolmogorov Test	Scale Parameter	Shape Parameter
TSP (1986)	lognormal	yes	4.317	0.381
SSI $PM_{10}$ (1986)	lognormal	yes	3.594	0.459
Beta Scattering Coefficient (1979-84)	66% lognormal	-	-	0.57-0.68

The shape parameter ( $\ln(\beta)$ ) for the preferred statistical distribution of SSI  $PM_{10}$  data (see Table 7-2) is comparable to that for TSP at Rocklea, 1986. The shape parameters for the two parameter lognormal distribution calculated using the inverse of wind speed over the period 1950 to 1984 (Simpson et al, 1987) range from 0.351 to 0.604. This encompasses both TSP (0.381) and  $PM_{10}$  (0.459) shape parameters and thereby supports assumption (2). It may, therefore, be possible to use the HEC modelling approach to predict at least inhalable particulate matter, measured as SSI  $PM_{10}$ , in much the same way as it has been done for TSP (see Figure 3-1). This would, however, require access to an appropriate emissions inventory.

Assumption (1) is acceptable for respirable particles, as measured by the beta

scattering coefficient, with 66% of the data sets between 1979 and 1984 being preferred lognormal<sup>1</sup>. It has been noted, however, that empirical assumptions different to assumption (2) may need to be made to determine an appropriate shape parameter for this distribution (Simpson et al, 1986).

## 7.5. DISCUSSION

An examination of health effects of particulate air pollution reveals that, while TSP is a good aggregate gravimetric measure of particulate air pollution, inhalable and respirable particulates are of primary health concern and therefore must be monitored separately. Consequently it must be accepted that there is a responsibility to control particles in this size range. Two fundamental aspects of the exposure-response relationship for inhalable and respirable particles are particle size and size dependent chemical composition. Due to potentially long retention times and the pulmonary exchange of chemicals in the alveolar region of the lung, chemical analysis of the respirable size fraction,  $<2.5\mu\text{m}$ , should be obtained if possible.

It should be acknowledged that this level of resolution has not been adopted by the USEPA. The development of standards for  $\text{PM}_{10}$  has been preferred. It is likely that ambient air quality goals will be similarly adopted in Australia in terms of  $\text{PM}_{10}$ . Although this measure may provide only part of the picture, with information on the respirable size fraction being largely lost through aggregation, it is a robust technique that utilises existing high-volume sampling techniques. If further resolution and access to size dependent chemical composition data in the respirable range is desired then techniques such as dichotomous sampling could be used.

Currently in Brisbane ambient data at this resolution are not available with SSI  $\text{PM}_{10}$  and nephelometers used to obtain information on inhalable and respirable particulates. As no mass is collected for the respirable particles a quantitative measure of composition in this fraction is limited. Consequently SEM micro-probe analysis has been used to identify the composition and potential health concerns of the respirable particles. Lead and sulphur are two compounds that are of specific health concern (Murray et al, 1987) and have been identified as constituents of respirable particles in the Brisbane urban area.

Ambient concentrations must also be related to emissions in order to identify

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<sup>1</sup> preferred lognormal, 1 preferred gamma but accepted as lognormal by the Kolmogorov statistic and 1 preferred gamma outright between 1979-84 (Simpson et al, 1986)

appropriate controls. The TSP HEC model has been shown to make this link relatively simply for TSP and provide acceptable results. Two approaches may be used to relate this model and modelling technique to inhalable and respirable particulates.

Firstly the relationship of TSP, and therefore TSP HEC predictions, to  $PM_{10}$  highlights some similarities in the response of these ambient measurements to source strengths and meteorological conditions. Beta scattering coefficients are less correlated with TSP. Predictions made by the TSP HEC model may therefore only be able to be related to inhalable particulates measured as SSI  $PM_{10}$  in Brisbane. To do this it must be assumed that the relationship holds throughout the Brisbane airshed. The predictions of ground level TSP concentration could then be related to  $PM_{10}$  through a constant. For Rocklea this constant was 0.50 which compares well with other estimates of this relationship (Rodes and Evans, 1985, Woodland, 1985). No relationship of this type has yet been identified between TSP HEC model predictions and respirable particles measured as beta scattering coefficients.

For the most reliable predictions, and in order to relate estimates of ambient ground level concentrations to emissions of inhalable and respirable particulates, the HEC modelling approach outlined in Figure 3-1 should be applied to these size ranges independently of TSP. This approach is first dependent upon satisfying the assumptions of the HEC approach. It has been found that these assumptions, necessary for the execution of the TSP HEC model, are valid for  $PM_{10}$ . Secondly, to execute the HEC model to predict  $PM_{10}$  ground level concentrations directly a modified emissions inventory would also be necessary.

It has been noted that the average difference between TSP and  $PM_{10}$  is approximately equivalent to the background crustal component for Rocklea. This warrants further investigation as it may be possible to simply remove anthropogenic crustal contributions from the emissions inventory (with background crustal sources already not included) and thereby approximate an emissions inventory for  $PM_{10}$ . It would have to be assumed that the remaining anthropogenic emissions were approximately less than  $10 \mu m$ . This may not be unreasonable as the particulate sources in this inventory are dominated by combustion processes from domestic, motor vehicle and industrial sources (Borowski, 1984). If this assumption is sufficiently valid then a relatively crude HEC model may be developed for  $PM_{10}$ . To fulfill the requirements of an appropriate emissions control strategy for both inhalable and respirable particulates, however, this inventory would have to be updated to include size and composition data.

The incorporation of chemical composition analyses together with the HEC model predictions, through techniques such as receptor modelling, would enable more accurate apportionment of source contributions. In the absence, however, of chemical composition data to link specific health effects to specific source types the HEC modelling approach may still provide a valuable tool for the development and implementation of a control strategy for inhalable particulates as SSI PM<sub>10</sub> in Brisbane.

## Chapter 8

# CONCLUSIONS

The objective of this study has been to produce an air quality model that can predict ground level concentrations of particulate pollution with an acceptable level of accuracy in an urban environment; these predictions must relate to both the standards that are used to define clean air and the emissions that have contributed to these concentrations. The model can then be used to assist in the development and implementation of control strategies for particulate air pollution where these concentrations are unacceptable.

The ATDL model has been found to satisfy this objective in terms of a recognised long term goal, the annual arithmetic mean concentration of  $90\mu\text{gm}^{-3}$  for TSP. This has only been possible by calibrating the model output using estimates of background crustal TSP from additional analyses conducted by the DNAAPC in 1985. A hybrid model will also produce sufficiently accurate estimates of TSP. This model provides output in terms of an entire frequency distribution for TSP and therefore any standard relating to that distribution (eg USEPA 24 hour maximum) can be used to help define acceptable air quality. In terms of the objectives of this study, however, this model only links ambient concentrations to total emissions and therefore does not provide sufficient detail to support source specific controls.

By using the hybrid approach, however, it has been possible to extend the calibrated ATDL model and produce the TSP hybrid emissions control (HEC) model. This has involved, first, determining the most appropriate statistical distribution for TSP data in Brisbane. Using wind speed data to provide a measure of the variation for that distribution (ie the shape parameter) the ATDL model output, as an annual arithmetic mean, is then related to an entire frequency distribution for predicted particulate concentrations. This allows emission controls to be linked through the ATDL model to violations of both long and short term exposure standards.

It has been shown that the TSP HEC model satisfies the objectives of this study by generally relating specific emissions to short and long term ambient standards

at an acceptable level of accuracy (ie to within a factor of 2). The model can therefore be used to assist in air quality control strategy development and implementation on a detailed level. Through the use of the hybrid modelling approach predictions can be made for an entire frequency distribution. This provides flexibility in the range of standards or goals that can be used to compare with model predictions and thereby define the acceptability of the air under investigation. Indeed, it has become clear that it is necessary to review both short and long term goals together (derived from the same frequency distribution) as controls to avoid violations of the long term annual arithmetic mean for TSP will not necessarily protect against the effects of short term exposure.

For most sites and years between 1979 and 1984 the TSP HEC model provides acceptable estimates, to within a factor of two, of mean and maximum ambient concentrations based upon the 1978/79 emissions inventory. The model may, therefore be applicable to current management strategies in Brisbane for particulate air pollution. While the addition of chemical composition data is important to produce an accurate prediction of TSP it is noteworthy that violations of current air quality goals do not occur using the ATDL model based upon anthropogenic emissions alone. It appears that the background crustal contributions, of limited health concern, are important in interpreting appropriate controls where TSP concentrations violate health goals.

The ATDL component of the TSP HEC model enables point and area source contributions to be distinguished from each other. This has allowed recommendations to be made with respect to specific sources that need to be controlled in Brisbane. Similarly, by hypothetically reducing emissions from motor cars by 24% the effect of the introduction of unleaded petrol on predicted arithmetic mean and maximum ground level concentrations has been estimated. It is concluded that both point and area source controls are necessary in Brisbane and that unleaded petrol will not be sufficient to reduce ambient TSP concentrations to within acceptable levels, as defined by the NHMRC and USEPA, at all sites.

The predictions made by the TSP HEC model are generally accurate to within a factor of 2. While this is acceptable for air quality models (Hanna, 1982) the conclusions drawn from these results should only be considered as guidelines to possible action. The decision makers must interpret these recommendations based on the control philosophy that embodies the social, economic and political issues relevant to the Brisbane airshed and community. The primary assumptions that accompany this uncertainty are that:

1. the ATDL model is valid for the prediction of the annual arithmetic mean;
2. the two parameter lognormal distribution is appropriate to represent TSP data in Brisbane; and,
3. a simple inverse relationship exists between percentile values of TSP and wind speed.

The lognormal assumption (2) has been found to be valid for most sites and years in Brisbane, Sydney, Melbourne, <sup>and, to a lesser extent,</sup> Adelaide and Canberra. If assumption (2) is invalid and gamma or Weibull distributions are more appropriate then overprediction and a conservative emissions control strategy is likely. Similarly the use of the inverse relationship in assumption (3) will result in over-estimation of TSP maxima for the sites examined in Sydney, Melbourne and Canberra. The absolute errors in estimating the annual 24 hour averaged maximum if these assumptions are invalid range up to 161% for Brisbane, Sydney, Melbourne and Adelaide<sup>1</sup>. Furthermore the areas of Brisbane, Sydney and Melbourne appear to be distinct from Canberra in terms of both assumptions (2) and (3) with absolute errors up to a factor of three estimated for this inland site.

Assumption (3) is weak as it is unlikely that variation in the wind speed data alone will be sufficient to explain the variance of particulate air pollution data under all situations. In fact the variance in wind speed data is greater than that experienced for monitored TSP concentrations. In support of this it has been found that the absolute errors in estimating the 365 day maximum for the lognormal distribution may be reduced if the geometric standard deviation derived from assumption (3) is replaced by the average geometric standard deviation for all TSP sites used in this study.

Results indicate that both assumptions may be useful as they would tend to produce a conservative control strategy for particulate emissions and that they may be generally applicable for urban areas where particulate emissions have little marked seasonal variability. It was also found that the use of a mean value for the geometric standard deviations of TSP concentrations, based on averaging  $\beta_{\text{TSP}}$  for all east and south east coast sites for all years, improved the model performance. It is clear that more work in this area is warranted. The results presented here provide only a rough guide to the potential applicability of the TSP HEC model.

Finally, it has been made clear that if the model predictions are to relate to

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<sup>1</sup> only assumption (2) has been tested for Adelaide due to the unavailability of wind speed data



both accepted standards and criteria for the protection of human health, then inhalable and respirable particles must be investigated. It may be possible to relate the HEC model to more "health relevant" inhalable particulate concentrations measured as  $PM_{10}$  in addition to TSP. SSI  $PM_{10}$  data have been shown to satisfy the assumptions of the TSP HEC model ((2) and (3) detailed earlier in this Chapter). In order to apply the HEC approach to  $PM_{10}$  data, however, an appropriate emissions inventory is required. It would be possible, given a complete emissions inventory or additional chemical composition data, for this HEC model to be successfully operated by regulatory authorities to control inhalable and respirable particulates. This would enable decision-makers to control both existing emissions and the introduction of new sources, relating to specific health concerns, in a given urban area in terms of both short and long term air quality goals.

In addition to the primary assumptions that have been outlined in this thesis a two caveats for this research should be presented. First, the assumption that the introduction of unleaded petrol will have reduced particulate emissions from motor vehicles by 24% in 1990 is very weak. This will be influenced greatly by socio-economic factors which help determine the rate at which new vehicles are introduced to the total fleet and the vehicle kilometers travelled by these cars. However, even if the reduction in emissions is inaccurate, this estimate provides a good example of how the TSP HEC model may be used to assist in control strategy development. Second, there is considerable uncertainty as to the topographic and meteorological variations throughout the Brisbane airshed. While the TSP HEC model appears to be valid for most of the monitored sites in Brisbane, it would be prudent to confirm this at other sites.

Further areas of research that are suggested by this work include:

- improving the geographic interpretation of the model through the inclusion of topographic data;
- modifying the emissions inventory to account for inhalable and respirable particulates;
- providing additional wind field data to account for localised variation in meteorological conditions; and,
- the use of statistical techniques to estimate confidence intervals for TSP HEC predictions.

## Appendix A

### Modified ATDL Model FORTRAN Code

*based extensively on code detailed by Hanna (1972).*

```

C      DESCRIPTION: ATMOSPHERIC TURBULENCE DIFFUSION MODEL TO BE USED
C      WITH BRISBANE EMISSIONS INVENTORY DATA FOR PARTICULATES,
C      AND COMBINED WITH CRES/AES-HYBRID MODEL OUTPUT PROVIDING
C      MEDIAN AND MAXIMUM VALUES FOR EACH GRID **ADJUSTED FOR
C      BACKGROUND TSP CONCENTRATIONS**
C      GERALD MILES ,SEPTEMBER 1986
C      CONSTRAINTS: GRID SIZE = 4KM BY 4KM.
C      VARIABLE LIST:      INPUT VARIABLES
C                          BETA - SHAPE PARAMETER (LOGNORMAL DISTRIBUTION)
C                          DX-GRID DISTANCE (M)
C                          BX-RURAL SOURCE STRENGTHS (UG/M**2.S)
C                          NR-NO. OF ROWS,INCLUDING 4 EXTRA ROWS AT TOP AND BOTTOM
C                          NC-NO. OF COLUMNS,INCLUDING 4 EXTRA COLUMNS AS FOR ROWS
C                          NO-NUMBER OF CORRECTIONS TO AREA SOURCE STRENGTHS
C                          NH-NO. OF EFFECTIVE SOURCE HEIGHT CLASSES
C                          NN-NO.OF STABILITY CLASSES
C                          RX(10)-PARAMETERS 'A' IN CALCULATION OF SIGMA Z
C                          PX(10)-PARAMETERS 'B' IN CALAULATION OF SIGMA Z
C                          U-WIND SPEED (M/S)
C                          F(16)-WIND DIRECTION FREQUENCY DISTRIBUTION (CLOCKWISE FROM
C                          NNE)
C                          ITIT1-DESCRIPTION OF AREA SOURCES
C                          ITIT2-DESCRIPTION OF POINT SOURCES
C                          ITIT3-DESCRIPTION OF CONCENTRATION PATTERNS
C                          ID(40)-ROW NO. OF AREA SOURCE CORRECTION
C                          JD(40)-COLUMN NO. OF AREA SOURCE CORRECTION
C                          ST(40)-AREA SOURCE CORRECTIONS (UG/M**2.S)
C                          S(40,40)-AREA SOURCE STRENGTHS (UG/M**2.S)
C                          H(20)-EFFECTIVE SOURCE HEIGHTS (M)
C                          NP(20)-NO. OF POINT SOURCES IN EACH HEIGHT CLASS
C                          IC(10,40)-ROW NO. OF POINT SOURCE
C                          JC(10,40)-COLUMN NO. OF POINT SOURCE
C                          QEC(10,40)-POINT SOURCE STRENGTH (UG/S)
C                          PROGRAM AND OUTPUT VARIABLES
C                          FU(9,9)-MATRIX OF DIRECTION FREQUENCY DISTRIBUTION DIVIDED
C                          BY WIND SPEED
C                          C(9,9)-MATRIX OF COEFFICIENTS FOR AREA SOURCES
C                          A(40,40)-CONCENTRATION DUE TO AREA SOURCES (UG/M**3)
C                          SS(9,9)-MATRIX OF COEFFICIENTS FOR CURRENT POINT SOURCES
C                          AA(40,40)-CONCENTRATION DUE TO CURRENT POINT SOURCES (UG/M)
C                          AC(40,40)-CONCENTRATION DUE TO ALL SOURCES (UG/M**3)
C                          RAT(40,40)-RATIO OF 'A' TO 'AC'
C                          S(40,40)-MATRIX OF RURAL SOURCE STRENGTHS
C                          D(40,40)-USED IN THE CALCULATION OF SIGMA Z
C                          QE(40,40)-CONTAINS VALUES FOR QEC(NI,K)
C                          AAB(40,40)-USED IN THE CALCULATION OF AC(I,J)
C                          ST(100)-CONTAINS VALUES OF ST(K)
C                          ALPHA(40,40)-MATRIX OF MEDIAN CONC. DUE TO ALL SOURCES
C                          AMAX(40,40)-MATRIX OF MAX. CONC. DUE TO ALL SOURCES
C
C      DIMENSION S(40,40),FU(40,40),C(40,40),A(40,40),QE(40,40),D(40,40),
C      $SS(40,40),AA(40,40),AC(40,40),IC(10,40),JC(10,40),QEC(10,40),
C      $ALPHA(40,40),AMAX(40,40),ID(40),JD(40),ST(100),RX(10),PX(10),

```

```

$BACK(40,40),H(20),NP(20),AAB(40,40),RAT(40,40),F(16)
C
  CHARACTER*80 ITIT1,ITIT2,ITIT3
C
C      FORMATTING FOR INPUT DATA
100  FORMAT(3F10.2,4I5)
105  FORMAT(8F10.4)
110  FORMAT(A80)
111  FORMAT(/ /2X,A80/)
163  FORMAT(16I5)
C
      INPUT DATA
      READ(21,100)BETA,DX,BX,NR,NC,NO,NH
      WRITE(6,100)BETA,DX,BX,NR,NC,NO,NH
      DO 300 I=1,NR
      DO 300 J=1,NC
300  S(I,J)=BX
      READ(21,163)NN
      READ(21,105)(RX(I),I=1,NN)
      READ(21,105)(PX(I),I=1,NN)
      READ(21,105)U
      READ(21,105)(F(I),I=1,16)
      READ(21,110)ITIT1
      READ(21,110)ITIT2
      READ(21,110)ITIT3
      READ(21,163)(ID(I),I=1,NO)
      READ(21,163)(JD(I),J=1,NO)
      READ(21,105)(ST(I),J=1,NO)
C
      REVIEW OF INPUT DATA
      WRITE(6,163)NN
      WRITE(6,105)(RX(I),I=1,NN)
      WRITE(6,105)(PX(I),I=1,NN)
      WRITE(6,105)U
      WRITE(6,105)(F(I),I=1,16)
      WRITE(6,110)ITIT1
      WRITE(6,110)ITIT2
      WRITE(6,110)ITIT3
      WRITE(6,163)(ID(I),I=1,NO)
      WRITE(6,163)(JD(I),J=1,NO)
      WRITE(6,105)(ST(I),J=1,NO)
      JI=NC-4
      IJ=NR-4
      DO 301 I=5,IJ
      READ(21,105)(S(I,J),J=5,JI)
      WRITE(6,105)(S(I,J),J=5,JI)
      DO 999 J=5,JI
      S(I,J)=S(I,J)/504.576
999  CONTINUE
301  CONTINUE
      READ(21,105)(H(I),I=1,NH)
      READ(21,163)(NP(I),I=1,NH)
      WRITE(6,105)(H(I),I=1,NH)
      WRITE(6,163)(NP(I),I=1,NH)
      DO 860 I=1,NH
      NNP=NP(I)
      READ(21,163)(IC(I,J),J=1,NNP)
      READ(21,163)(JC(I,J),J=1,NNP)
      READ(21,*)(QEC(I,J),J=1,NNP)
      WRITE(6,163)(IC(I,J),J=1,NNP)
      WRITE(6,163)(JC(I,J),J=1,NNP)
      WRITE(6,*)(QEC(I,J),J=1,NNP)
      DO 998 J=1,NNP
      QEC(I,J)=QEC(I,J)*31709.792
998  CONTINUE
860  CONTINUE
C
      WIND ROSE OUTPUT
      WRITE(20,794)(F(I),I=1,16)
794  FORMAT(1H1,2X,9HWIND ROSE/(8F10.4))
      DO 789 I=1,9
      DO 789 J=1,9
789  FU(I,J)=0.0

```

```

C          'E' BECOMES THE INVERSE OF WIND SPEED
E=1.0/U
FU(5,5)=E
C          THE NEXT 30 LINES DETERMINE THE MATRIX OF FREQUENCY/WIND SPEED
DO 790 I=1,4
FU(5-I,5+I)=F(2)*E
FU(5+I,5+I)=F(6)*E
FU(5+I,5-I)=F(10)*E
FU(5-I,5-I)=F(14)*E
FU(5-I,5)=F(16)*E
FU(5,5+I)=F(4)*E
FU(5+I,5)=F(8)*E
790 FU(5,5-I)=F(12)*E
FU(4,5)=FU(4,5)+E*(F(1)+F(15))
FU(5,6)=FU(5,6)+E*(F(3)+F(5))
FU(6,5)=FU(6,5)+E*(F(7)+F(9))
FU(5,4)=FU(5,4)+E*(F(11)+F(13))
DO 791 I=1,2
FU(1+I,6)=F(1)*E
FU(4,6+I)=F(3)*E
FU(6,6+I)=F(5)*E
FU(6+I,6)=F(7)*E
FU(6+I,4)=F(9)*E
FU(6,1+I)=F(11)*E
FU(4,1+I)=F(13)*E
791 FU(1+I,4)=F(15)*E
FU(1,7)=F(1)*E
FU(3,9)=F(3)*E
FU(7,9)=F(5)*E
FU(9,7)=F(7)*E
FU(9,3)=F(9)*E
FU(7,1)=F(11)*E
FU(3,1)=F(13)*E
FU(1,3)=F(15)*E
C          THE NEXT 8 LINES OUTPUT THE FREQUENCY/WIND SPEED MATRIX, GRID NETWORK
C          DIMENSIONS, EFFECTIVE SOURCE HEIGHTS, RURAL SOURCE STRENGTHS AND WIND SPEED
WRITE(20,9)((FU(I,J),J=1,9),I=1,9)
9  FORMAT(/2X,37HINPUT WIND SPEED AND FREQUENCY MATRIX/(9F10.5))
WRITE(20,470)NR,NC,NO,NH
470 FORMAT(/2X,9HNO. ROWS=,I5,3X,12HNO. COLUMNS=,I5,3X,4HNO.=,I5,3X,
$21HNO EFFECT SOURCE HTS=,I5)
WRITE(20,10)DX,BX,U
10  FORMAT(/2X,9HDX IN M =,F6.0,3X,23HRURAL SOURCE STRENGTHS=,F5.1,
$3X,11HWIND SPEED=,F10.3,5HM/SEC)
DO 600 I=1,NR
DO 600 J=1,NC
600 QE(I,J)=0.0
DO 167 K=1,NO
I=ID(K)
J=JD(K)
167 S(I,J)=ST(K)
C          OUTPUT OF AREA SOURCE DESCRIPTION
WRITE(20,*)ITIT1
DO 203 I=1,NR
WRITE(20,201)(S(I,J),J=1,NC)
201 FORMAT(10F10.2)
203 CONTINUE
DO 162 II=1,NN
C          SETS CURRENT VALUES OF 'A' AND 'B' FOR THE CALCULATION OF SIGMA Z
R=RX(II)
B=PX(II)
WRITE(20,119)R,B
119 FORMAT(1H1,' NEW SET OF POWER LAW PARAMETERS FOR SIGMA Z'/3X,
$2HR=,F5.3,3X,2HB=,F5.3)
IF(NO)160,160,150
150 CONTINUE
DO 1 I=1,9
DO 1 J=1,9
1  C(I,J)=0.0
BB=((DX/2.0)**(1.0-B))/(R*(1.0-B))

```

```

CC=9.0**(1.0-B)-7.0**(1.0-B)
DD=0.8*BB*CC
DO 2 J=1,9,2
C(1,J)=DD*FU(I,J)
2 C(9,J)=DD*FU(9,J)
DO 3 I=3,7,2
C(I,1)=DD*FU(I,1)
3 C(I,9)=DD*FU(I,9)
CC=7.0**(1.0-B)-5.0**(1.0-B)
DD=0.80*BB*CC
C(8,8)=DD*FU(8,8)
C(2,2)=DD*FU(2,2)
C(2,8)=DD*FU(2,8)
C(8,2)=DD*FU(8,2)
DO 4 J=4,6
C(2,J)=DD*FU(2,J)
4 C(8,J)=DD*FU(8,J)
DO 5 I=4,6
C(I,2)=DD*FU(I,2)
5 C(I,8)=DD*FU(I,8)
CC=5.0**(1.0-B)-3.0**(1.0-B)
DD=0.80*BB*CC
DO 6 J=3,7
C(3,J)=DD*FU(3,J)
6 C(7,J)=DD*FU(7,J)
DO 7 I=4,6
C(I,3)=DD*FU(I,3)
7 C(I,7)=DD*FU(I,7)
CC=3.0**(1.0-B)-1.0
DD=0.80*BB*CC
DO 8 J=4,6
C(4,J)=DD*FU(4,J)
8 C(6,J)=DD*FU(6,J)
C(5,4)=DD*FU(5,4)
C(5,6)=DD*FU(5,6)
C(5,5)=0.80*BB*FU(5,5)
WRITE(20,11)((C(I,J),J=1,9),I=1,9)
11 FORMAT(/2X,39HMATRIX OF COEFFICIENTS FOR AREAS SOURCES/(9F10.5))
DO 500 I=5,IJ
DO 500 J=5,JI
A(I,J)=0.0
DO 500 K=1,9
DO 500 L=1,9
500 A(I,J)=C(K,L)*S(I-5+K,J-5+L)+A(I,J)
WRITE(20,95)
95 FORMAT(/2X,34HCONCENTRATIONS FROM GROUND SOURCES)
WRITE(20,111)ITIT3
DO 690 I=5,IJ
DO 690 J=5,JI
A(I,J)=A(I,J)+42.0
690 CONTINUE
DO 701 I=5,IJ
WRITE(20,700)(A(I,J),J=5,JI)
700 FORMAT(10F10.4)
701 CONTINUE
160 IF(QEC(1,1))162,162,161
161 CONTINUE
WRITE(20,202)
202 FORMAT(/2X,16HELEVATED SOURCES)
WRITE(20,111)ITIT2
DO 899 I=5,IJ
DO 899 J=5,JI
899 AAB(I,J)=0.0
DO 880 NI=1,NH
DO 874 KL=1,NR
DO 874 KM=1,NC
874 QE(KL,KM)=0.0
WRITE(20,871)H(NI)
871 FORMAT(2X,26HEFFECTIVE SOURCE HEIGHT = ,F10.3)
WRITE(20,872)

```

```

872  FORMAT(10X,6HROW NO,3X,9HCOLUMN NO,4X,21HEMISSION(UGM PER SEC))
      NNP=NP(NI)
      WRITE(20,873)((IC(NI,K),JC(NI,K),QEC(NI,K)),K=1,NNP)
873  FORMAT(21I2,F20.5)
      DO 164 K=1,NNP
        I=IC(NI,K)
        J=JC(NI,K)
164   QE(I,J)=QEC(NI,K)
        DO 81 L=1,9
          DO 81 K=1,9
81     D(L,K)=0.0
        XY=H(NI)**2.0/2.0
        P=EXP(-XY/(R**2.0*((DX/2.0)**(2.0*B))))
        PA=R*((DX/2.0)**(B+1.0))
        XINT=0.
        DDR=0.
        XX2=DX/2.
        DO 948 IJJ=1,100
          IF(IJJ-10)952,952,953
952   DR=100.
          GO TO 954
953   DR=1000.
954   DDDR=DDR+DR/2.
          DDR=DDR+DR
          XYZ=XY/(R**2.0*(DDDR**2.0*B))
          IF(XYZ-20.0)960,960,961
960   XINT=XINT+DR*EXP(-XYZ)/(DDDR**B)
961   CONTINUE
          IF(XX2-DDR)955,955,948
948   CONTINUE
955   CONTINUE
          D(5,5)=XINT/(8.0*R*XX2**2.0)
          DO 71 L=1,4
            E=2*L
            EF=E**2.0*B
            XF=1.4*E
            FE=XF**2.0*B
            Z=P**2.0/(EF*(PA*(E**2.0*(B+1.0))))
            IF(L-1)950,950,949
949   D(5,5+L)=Z
            D(5,5-L)=Z
            D(5+L,5)=Z
            D(5-L,5)=Z
            GO TO 951
950   D(5,6)=Z/3.
            D(5,4)=Z/3.
            D(6,5)=Z/3.
            D(4,5)=Z/3.
951   Z=P**2.0/(FE*(PA*(XF**2.0*(B+1.0))))
            D(5+L,5+L)=Z
            D(5+L,5-L)=Z
            D(5-L,5+L)=Z
71   D(5-L,5-L)=Z
            DO 72 L=1,2
              QQ=2*L+2
              QG=1.1*QQ
              G=QG**2.0*B
              Z=P**2.0/(G*(PA*(QG**2.0*(B+1.0))))
              D(4-L,6)=Z
              D(4-L,4)=Z
              D(6+L,4)=Z
              D(6+L,6)=Z
              D(4,4-L)=Z
              D(6,4-L)=Z
              D(4,6+L)=Z
              D(6,6+L)=Z
72   Z=P**2.0*(2.0*B)/(PA*(10.0**2.0*(B+1.0)))
            D(1,3)=Z
            D(1,7)=Z
            D(3,1)=Z

```

```

D(3,9)=Z
D(7,1)=Z
D(7,9)=Z
D(9,3)=Z
D(9,7)=Z
DO 91 K=1,9
DO 91 L=1,9
91 SS(K,L)=2.04*D(K,L)*FU(K,L)
WRITE(20,92)((SS(I,J),J=1,9),I=1,9)
92 FORMAT(/2X,33HCOEFFICIENTS FOR ELEVATED SOURCES/(9E13.6))
DO 510 I=5,IJ
DO 510 J=5,JI
AA(I,J)=0.0
DO 410 K=1,9
DO 410 L=1,9
ADD=SS(K,L)*QE(I-5+K,J-5+L)
AA(I,J)=AA(I,J)+ADD
410 AAB(I,J)=AAB(I,J)+ADD
510 CONTINUE
WRITE(20,96)H(NI)
96 FORMAT(/2X,50HCONCENTRATIONS FROM ELEVATED SOURCES WITH HEIGHT =,
$F10.3)
WRITE(20,111)ITIT3
DO 702 I=5,IJ
WRITE(20,700)(AA(I,J),J=5,JI)
702 CONTINUE
880 CONTINUE
IF(NO)166,165,166
166 CONTINUE
IF(NH)169,165,169
169 CONTINUE
DO 520 I=5,IJ
DO 520 J=5,JI
AC(I,J)=A(I,J)+AAB(I,J)
ALPHA(I,J)=AC(I,J)/(EXP(0.5*(LOG(BETA)**2)))
AMAX(I,J)=ALPHA(I,J)*BETA**2.94
520 RAT(I,J)=A(I,J)/AC(I,J)
WRITE(20,97)
97 FORMAT(2X,31HCONCENTRATIONS FROM ALL SOURCES)
WRITE(20,111)ITIT3
DO 703 I=5,IJ
WRITE(20,700)(AC(I,J),J=5,JI)
703 CONTINUE
WRITE(20,890)
890 FORMAT(2X,66H RATIO OF CONCENTRATION DUE TO AREA SOURCES TO TOTAL
$CONCENTRATION)
DO 704 I=5,IJ
WRITE(20,700)(RAT(I,J),J=5,JI)
704 CONTINUE
WRITE(20,891)
891 FORMAT(2X,' MEDIAN CONCENTRATIONS DUE TO ALL SOURCES')
DO 705 I=5,IJ
WRITE(20,700)(ALPHA(I,J),J=5,JI)
705 CONTINUE
WRITE(20,892)
892 FORMAT(2X,' MAXIMUM CONCENTRATIONS DUE TO ALL SOURCES')
DO 706 I=5,IJ
WRITE(20,700)(AMAX(I,J),J=5,JI)
706 CONTINUE
165 CONTINUE
162 CONTINUE
STOP
END

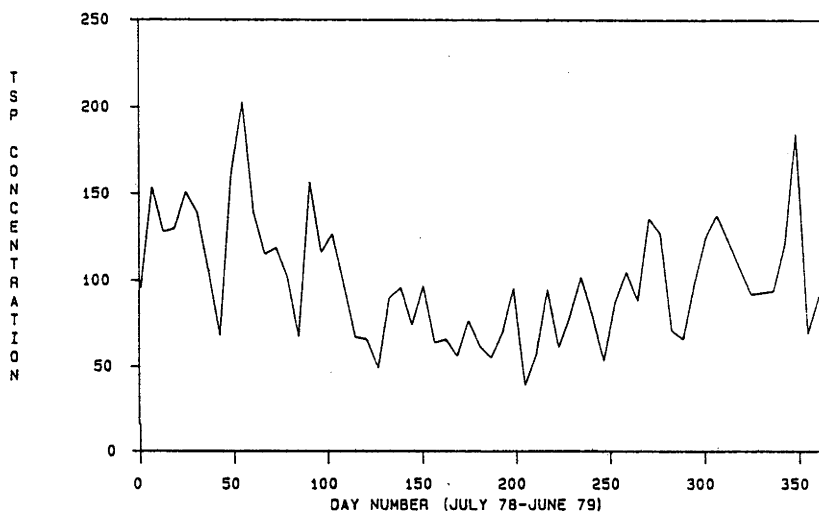
```

## Appendix B

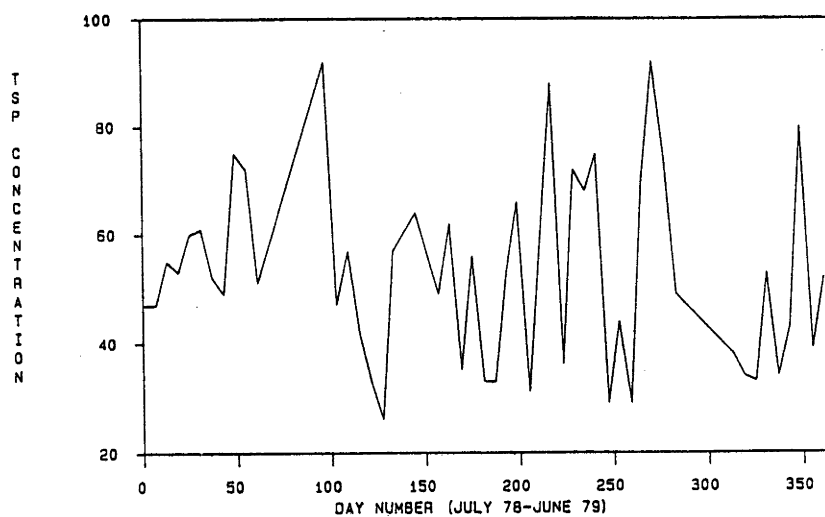
### Seasonality of TSP Data

The following Figures B-1 to B-6 represent the seasonal variation that was demonstrated at the TSP sites in Brisbane during 1978/79.

**Figure B-1:** Annual variation in TSP concentrations ( $\mu\text{gm}^{-3}$ ) at the Woolloongabba site 1978/79

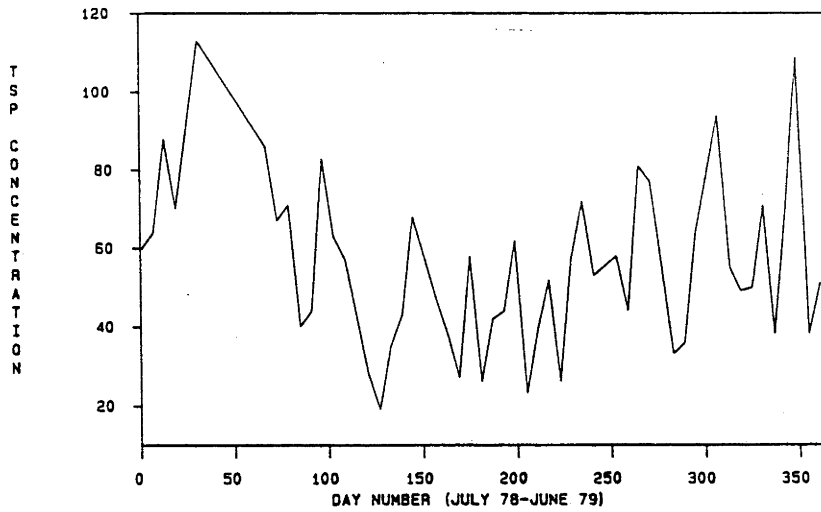


**Figure B-2:** Annual variation in TSP concentrations ( $\mu\text{gm}^{-3}$ ) at the Petrie Bight site 1978/79

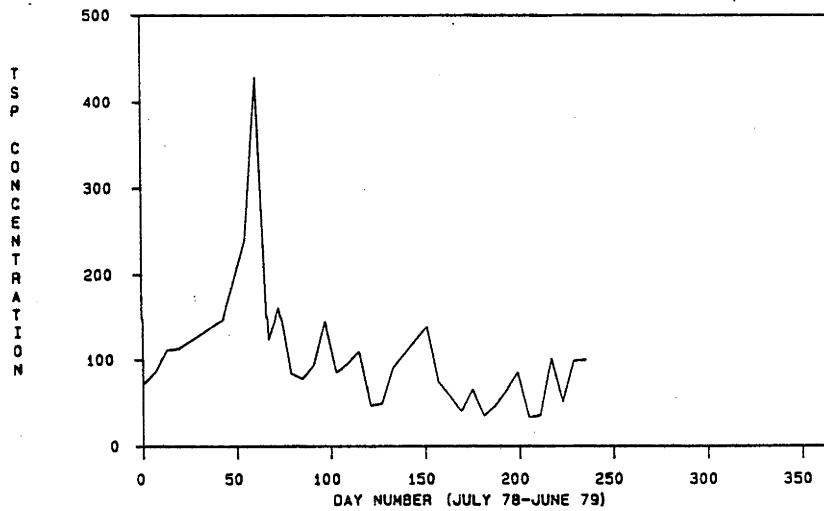




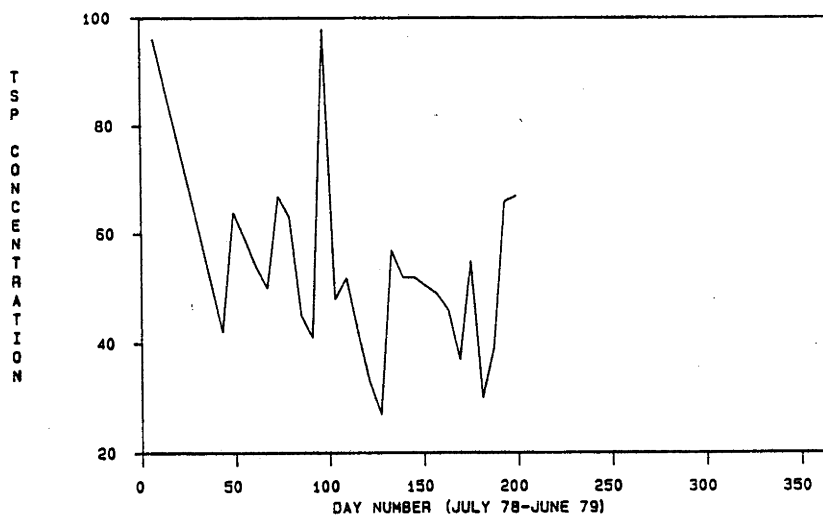
**Figure B-3:** Annual variation in TSP concentrations ( $\mu\text{gm}^{-3}$ ) at the Hamilton site 1978/79



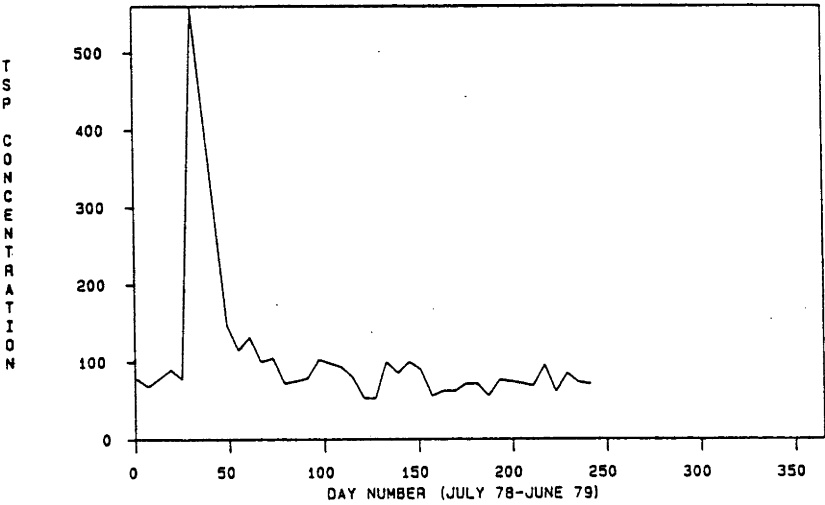
**Figure B-4:** Annual variation in TSP concentrations ( $\mu\text{gm}^{-3}$ ) at the QIT site 1978/79



**Figure B-5:** Annual variation in TSP concentrations ( $\mu\text{gm}^{-3}$ ) at the South Brisbane site 1978/79



**Figure B-6:** Annual variation in TSP concentrations ( $\mu\text{gm}^{-3}$ ) at the Normanby site 1978/79

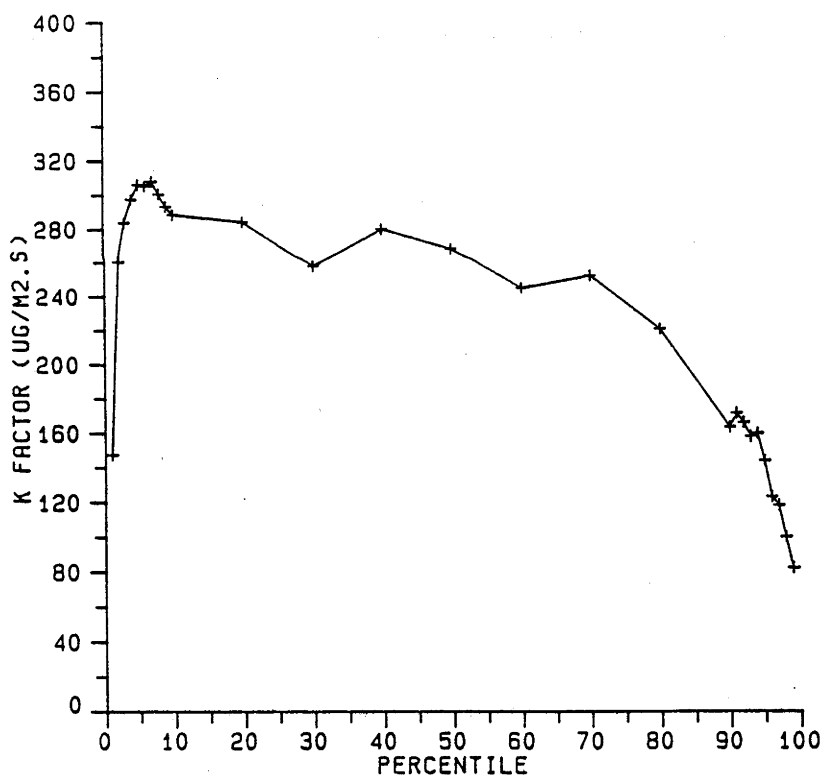


## Appendix C

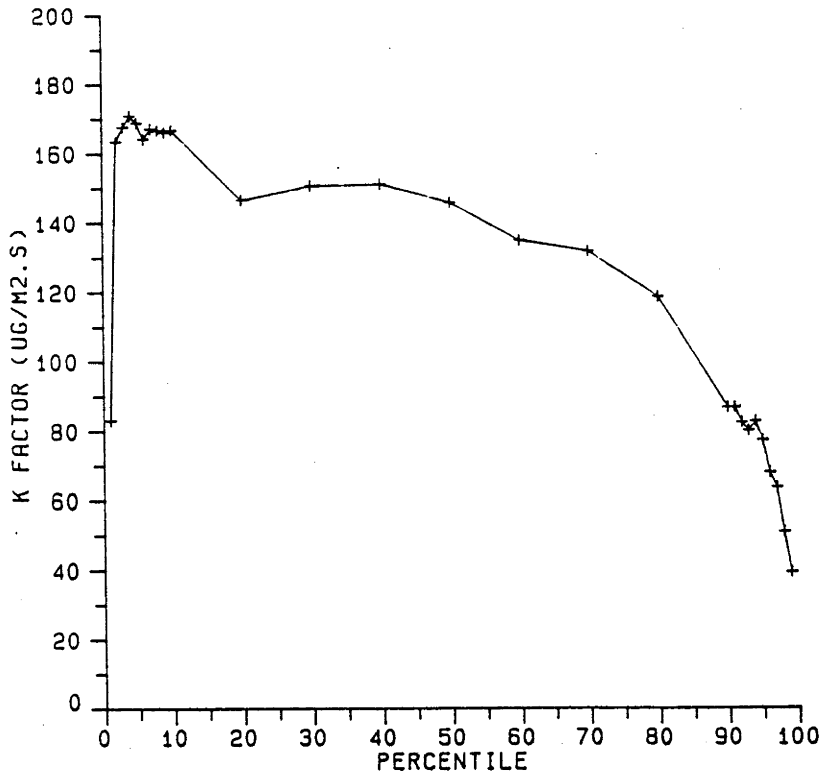
### Validity Of The Empirical Assumption

The following Figures C-1 to C-6 examine the constancy of  $K$ , from equation (3.5), over the range of percentiles for each site. The variation in the constancy of  $K$  for the sites of QIT, South Brisbane and Normanby may be explained in part by the incomplete records for TSP at these sites during 1978/79 (see Appendix B).

**Figure C-1:**  $K$  factors *versus* percentiles at the Woolloongabba site for TSP, 1978/79



**Figure C-2:** K factors *versus* percentiles at the Petrie Bight site for TSP, 1978/79



**Figure C-3:** K factors *versus* percentiles at the South Brisbane site for TSP, 1978/79

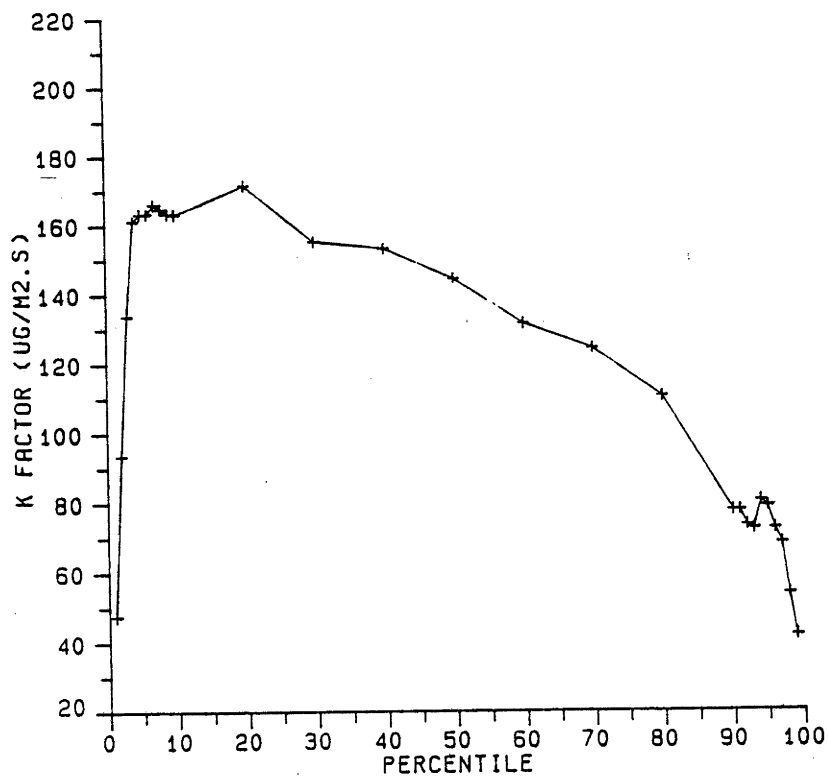


Figure C-4: K factors *versus* percentiles at the Normanby site for TSP, 1978/79

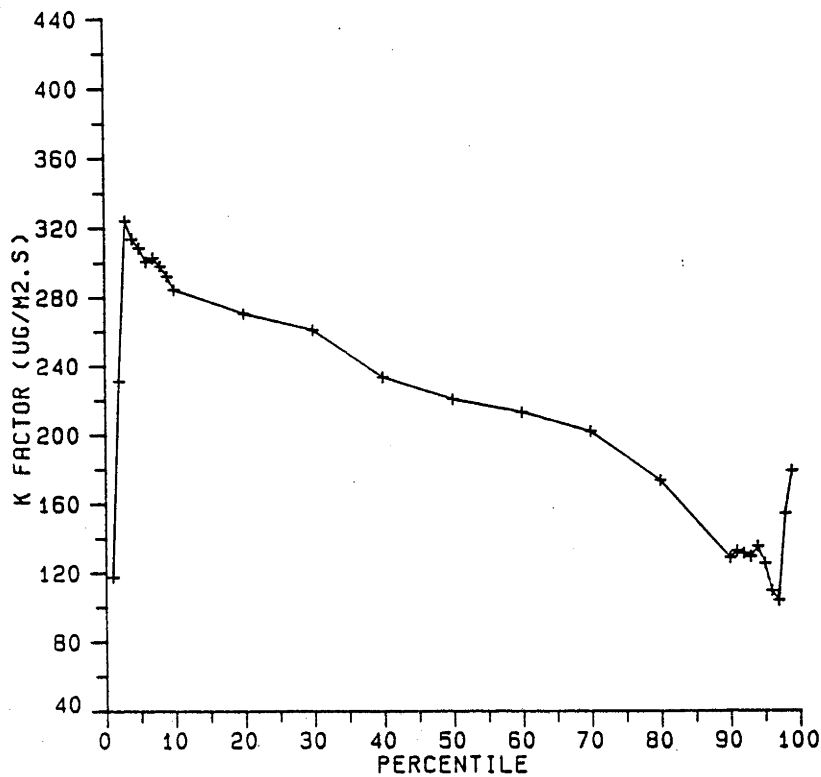


Figure C-5: K factors *versus* percentiles at the QIT site for TSP, 1978/79

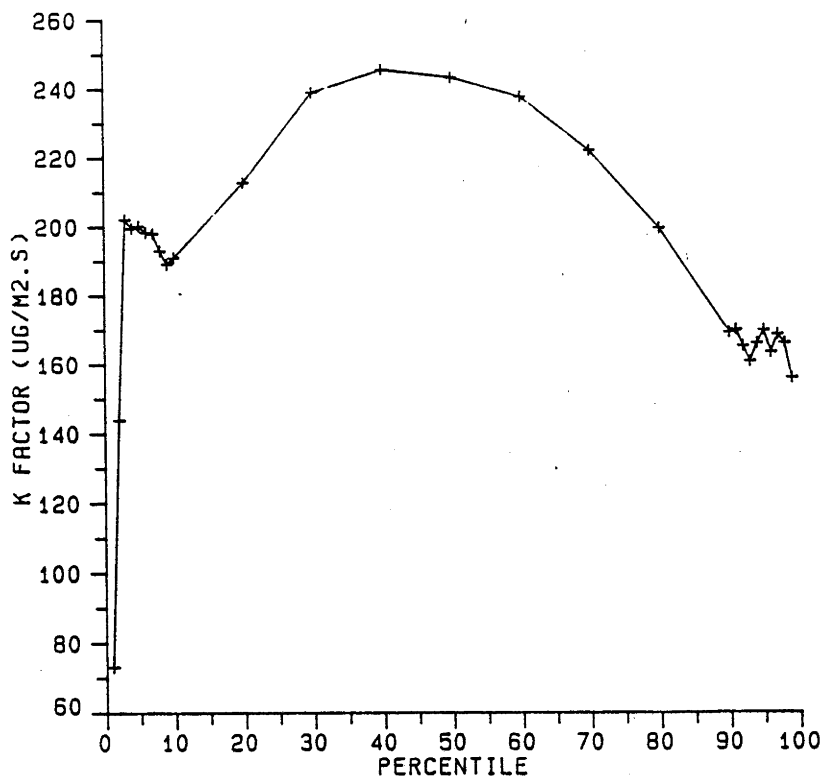
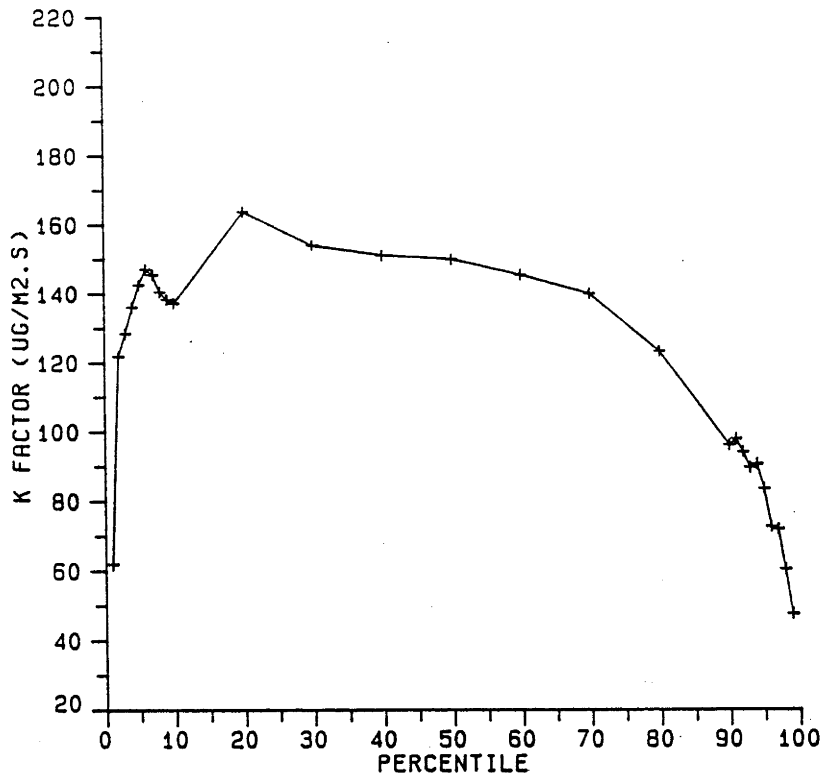


Figure C-6: K factors *versus* percentiles at the Hamilton site for TSP, 1978/79



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